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Procedure for automated low uncertainty assessment of empty cavity mode frequencies in Fabry-Pérot cavity based refractometry

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Abstract: A procedure for automated low uncertainty assessment of empty cavity mode frequencies in Fabry-Pérot cavity based refractometry that does not require access to laser frequency measuring instrumentation is presented. It requires a previously well-characterized system regarding mirror phase shifts, Gouy phase, and mode number, and is based on the fact that the assessed refractivity should not change when mode jumps take place. It is demonstrated that the procedure is capable of assessing mode frequencies with an uncertainty of 30 MHz, which, when assessing pressure of nitrogen, corresponds to an uncertainty of 0.3 mPa.

1. Introduction

Fabry-Pérot cavity (FPC) based refractometry is a technique that can be used for low uncertainty assessments of the refractivity of gases. By simultaneously measuring the temperature, and by using the Lorentz-Lorenz equation and an equation of state, it is also possible to assess pressure with high accuracy. With the latest revision of the SI-system, in which the Boltzmann constant was given a fixed value, it can even, via the use of ab initio quantum calculations of molar polarizabilities and relevant virial coefficients, provide an attractive path to a realization of the Pascal [1,2]. Since such a realization does not comprise any mechanical actuator, but instead depends on gas parameters, it can potentially decrease uncertainties and shorten calibration chains [3–12].

The basics of FPC-based refractometry relies on the fact that any given mode of a FPC will, when the cavity is filled with gas, shift its frequency by an amount that is given by the refractivity of the gas. Since the frequency of a cavity mode is normally assessed as the frequency of laser light that is locked to the mode, the refractivity (and thereby the molar density and pressure) of the gas is often practically assessed in terms of a shift in the frequency of the laser light when gas is let into (or evacuated from) a cavity.

Although assessments of refractivity involve measurements of the frequency of the laser light used and its shift when the cavity is filled with gas, they are also, in general, affected by a number of physical properties of the cavity, predominately its distortion when it is exposed to gas, the phase shift that the light experiences upon reflection from distributed Bragg reflector (DBR) equipped mirrors at their front facets, the spatial shape of its longitudinal modes (through the Gouy phase), and drifts in the length of the empty cavity [13].

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When a well-characterized FPC is used for assessment of refractivity (i.e. one whose physical properties have been assessed with low uncertainties) and when appropriate means have been taken to alleviate the influence of drifts of the cavity length [14], assessments of refractivity under low refractivity conditions are often limited by the ability to assess the frequency of the empty cavity mode addressed.

The assessment of the latter can, in turn, be affected by entities such as the ability to assess the frequency of the laser light, the locking of the laser to the cavity mode addressed, and the ability to provide appropriate conditions for an "empty" cavity assessment (i.e. a sufficiently low residual gas pressure). Since there are techniques available to lock lasers to cavity modes with high accuracy (to within Hz or better [15–18]), and there are means to provide appropriate conditions for "empty" cavity assessments (i.e. sufficiently low residual gas pressures), their influence on the assessment of the frequency of the empty cavity mode addressed can often be made inferior to that of the assessments of the frequency of the laser light. This implies that when a well-characterized FPC is used together with an adequate procedure for reduction of the influence of cavity drifts, FPC-based refractometry can, in practice, be limited by the ability to assess the frequency of the laser light.

Although there are techniques available to assess the frequency of laser light with low (sub-ppm) uncertainty, such instrumentation may not always be accessible during refractivity assessments. This is particularly the case when using transportable refractometry systems. Even if accessible, these instruments can demand complex and time-consuming measurement procedures, as is the case when employing a frequency comb. This means that such instrumentation is not always the most appropriate means to deal with this limitation.

As an alternative means to address the limited ability to assess the frequency of the laser light, this paper presents a procedure for how to assess, in a swift manner, the frequency of a laser addressing a mode in a well-characterized FPC-system that does not require access to any laser frequency measuring instrumentation. In particular, it addresses the conditions under which such a system can provide assessments of refractivity in the low refractivity regime [19,20] with a refractivity-independent uncertainty of 2.7×10^{-12}, henceforth referred to as the benchmark (BM) uncertainty and denoted $\delta(n-1)_{BM}$, which, for nitrogen at room temperature, corresponds to a pressure of 1 mPa.

The procedure requires that the system is well-characterized with respect to its physical properties, in particular with respect to the phase shifts of the light that take place at the front facets of the mirrors upon reflection (which is related to the mirror penetration depth) and the Gouy phase, and that the number of the mode addressed has been assessed [21], and makes use of an assessment of the free-spectral-range (FSR) of the cavity, i.e. the shift in frequency that takes place when a mode jump is induced

The organization of the paper is as follows: After a short review of commonly used expressions for the frequency of a FP-cavity mode and the refractivity assessed by the use of a dual frequency measurement methodology (in section 2), the paper identifies (in section 3, together with Supplement 1, the major contributions to the uncertainties in the assessments of refractivity under low refractivity conditions. Based on an analysis of these, it then defines the requirements that such a refractometer can provide assessments of refractivity below the benchmark, and shows that a well characterized FPC-based refractometer often can be limited by the uncertainty of the assessment of the frequency of the laser light when addressing an empty cavity [22].

Based on this, we then present, in section 4, a procedure for how the frequency of the laser light in a well-characterized FPC-system, even without access to any laser frequency measuring instrumentation, can be repeatedly and automatically assessed, in a swift and convenient manner, with such a low uncertainty that it makes possible assessments of refractivity down to the benchmark condition.

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Section 5 gives a brief description of the experimental setup while section 6 presents some results based on the presented procedure. Simultaneous assessments of the laser frequency performed by a highly accurate frequency comb confirm that the presented procedure has the ability to assess the laser frequency with the claimed accuracy.

2. Theory

2.1. Mode frequencies

As has been shown by Silander et al. [23], for the case with a cavity constructed by two mirrors with identical coating, the frequency of the m^{th} longitudinal cavity mode of a FP cavity exposed to a gas with an index of refraction of n can be expressed as

$$\nu = \frac{cm\left(1 + \frac{\Theta_G}{\pi m} + \frac{n\gamma_c}{m}\right)}{2n\left(L_0 + \delta L + 2L_{\tau,s}\right)}, \tag{1}$$

where c is the speed of light, m is an integer representing the number of the longitudinal mode the laser addresses, defined by Eq. (9) in Silander et al. [23], and Θ_G is the single pass Gouy phase. γ_c is a material-dependent entity related to the phase shift of the light at the front facet of the mirrors upon reflection, defined as $\frac{2\tau_c(n)\nu_c}{n}$, where, in turn, $\tau_c(n)$ is the group delay of a pulse of reflected light (defined as the time delay a narrow-band light pulse experiences upon reflection), and ν_c is the center frequency of the mirror coating [24], L_0 is the distance between the front facets of the DBRs coatings of the two mirrors when the cavity is evacuated, δL is the change in length of the cavity due to pressure induced deformation, and $L_{\tau,s}$ is the frequency penetration depth of the mirror coatings, defined as $\frac{c\tau_c(n)}{2n}$ [23,25].

2.2. Assessment of refractometry in FPC-based refractometry by the use of the single frequency measurement methodology

It is, in principle, possible to assess the refractivity, $n - 1$, of a gas directly from Eq. (1) by measuring the frequency, ν, of a given mode, m, of a well-characterized cavity (i.e. a cavity in which all physical properties, i.e. the Θ_G, γ_c, L_0, δL, and $L_{\tau,s}$ are known) when the cavity contains the gas addressed. However, as is further explicated below, this approach, below referred to as the "single frequency measurement" methodology, is not the most suitable means to assess gas refractivity. The reason is that it requires that both the length of the cavity and the laser frequency are assessed with such high accuracies that low uncertainty assessments of refractivity become extraordinarily challenging.

2.3. Assessment of refractometry in FPC-based refractometry by the use of the dual frequency measurement methodology

Instead, it has been found more suitable (and advantageous) to assess gas refractivity by a methodology, henceforth referred to as the "dual frequency measurement" methodology, that addresses the difference between the frequencies of the cavity modes addressed under two situations, one when the cavity is empty, denoted ν_0, and one when it is filled with the gas whose refractivity is to be assessed, $\nu(n-1)$, for simplicity henceforth denoted ν, carried out in (preferably rapid) succession.

Denoting this difference in frequency by $\Delta\nu$ (defined as $\nu_0 - \nu$), it is possible, as is shown by Silander et al. [23], to express (with a minimum of approximations, which are on the relative 10^{-9} to low 10^{-8} level) the refractivity of the gas in the cavity in terms of measurable and quantifiable

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quantities and material parameters as

$$n - 1 = \frac{\frac{\Delta\nu}{\nu_0}(1 + \frac{\Theta_G}{\pi m_0} + \frac{\gamma_c}{m_0}) + \frac{\Delta m}{m_0}}{1 - \frac{\Delta\nu}{\nu_0}(1 + \frac{\Theta_G}{\pi m_0} + \frac{\gamma_c}{m_0}) + \frac{\Theta_G}{\pi m_0} + n\varepsilon'}, \tag{2}$$

where Δm is the number of modes the laser has jumped during the filling (or emptying) of the cavity and m_0 is the number of the mode addressed in an empty cavity. We have here also introduced ε' as the refractivity-normalized relative elongation of the length of the cavity due to the presence of the gas, defined as $\frac{\delta L}{L_0'}\frac{1}{n-1}$, where, in turn, L_0' is the length of the cavity under vacuum conditions (thus when not being exposed to any pressure induced cavity distortion) as experienced by the light when its frequency is being altered, as for example takes place when the cavity is filled with (or emptied of) gas, given by $L_0 + 2L_{\tau,s}$ [25,26].

Although $\Delta\nu$ in principle can be assessed as the difference in the frequency of the laser addressing the measurement cavity when the cavity is empty and filled with gas by a frequency measuring instrumentation, it is more convenient to assess it as a shift of the beat frequency between two lasers, a measurement and a reference laser. An advantage of the latter is that the beat frequency can be conveniently and automatically assessed by a frequency counter with Hz rates, which is significantly faster than what the frequency of light can be assessed by any frequency measuring instrumentation. We will therefore, henceforth, assume that $\Delta\nu$ is assessed as the shift of a beat frequency measured by the use of a frequency counter. This implies that the $\Delta\nu$ entity henceforth will be referred to as the shift in the beat frequency.

2.4. Comparison between the abilities of the dual and the single frequency measurement methodologies to assess refractivity

The advantage of the "dual frequency measurement" methodology with respect to the "single frequency measurement" one is that the assessment of the shift in the beat frequency, as, for example, is done in the gas modulation refractometry (GAMOR) methodology, can be made significantly faster than separate assessments of L_0 and ν. As is further discussed below, this makes assessments made by the "dual frequency measurement" methodology, as compared to assessments made by the "single frequency measurement" one, less susceptible to drifts of the length of the cavity [27]. It also implies that such assessments benefit from more relaxed conditions of various physical properties, which significantly facilitates low-uncertainty assessments of refractivity (and thereby molar density and pressure).

3. Estimates of the requirements to assess refractivity within the benchmark

3.1. Single frequency measurement methodology

For the case when the single frequency measurement methodology is being used, for which the assessments are based directly on Eq. (1), it can be shown that the uncertainty in an assessment of refractivity in the low refractivity regime from an FPC system that is well characterized with respect to its physical properties and the number of the mode the laser addresses during the assessment is known, $\delta(n-1)$, is mainly given by the relative uncertainties in the empty cavity length, $\frac{\delta(L_0)}{L_0}$, and the frequency, $\frac{\delta(\nu)}{\nu}$, as

$$\delta(n-1) = \delta(n) = \sqrt{\left[\frac{\delta(L_0)}{L_0}\right]^2 + \left[\frac{\delta(\nu)}{\nu}\right]^2}, \tag{3}$$

where the $\delta(L_0)$ and $\delta(\nu)$ are the uncertainties in the estimates of the empty cavity length and the cavity mode frequency at the time instant when the refractivity assessment is carried out, respectively. This shows that a necessary condition to reach an uncertainty of refractivity given

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by the benchmark, i.e. a $\delta(n-1)_{BM}$ of 2.7×10^{-12}, is that the relative uncertainties in both the empty cavity length, i.e. the $\frac{\delta(L_0)}{L_0}$, and in the frequency, i.e. the $\frac{\delta(\nu)}{\nu}$, are assessed (or known) to at least this level. This implies that for a cavity with a length of 0.2 m, its empty cavity length needs to be known within half a pm (0.5×10^{-12} m), while, for a laser wavelength of 1.5 µm (for which ν_0 is 2×10^{14} Hz), the cavity mode frequency needs to be assessed with an uncertainty of 540 Hz.

It is non-trivial to assess these two entities to such degrees, in particular since the former additionally is affected by drifts that can take place between the time instants when the system was characterized with respect to its length and when the refractivity is assessed. Hence, despite the fact that it can be estimated by the use of extrapolation, it is truly challenging to assess refractivity with an uncertainty down to the benchmark by use of the single frequency measurement methodology.

3.2. Dual frequency measurement methodology

3.2.1. General expression for the uncertainty in the assessed refractivity

As can be concluded from a scrutiny of Eq. (2), when the dual frequency measurement methodology is being utilized, the corresponding uncertainty in the assessed refractivity from an FPC system in the low refractivity regime that is well characterized with respect to its physical properties [28], $\delta(n-1)$ can be expressed as

$$\delta(n-1) = \sqrt{\left[\frac{\delta(\Delta\nu)}{\nu_0}\right]^2 + \left[\frac{\Delta\nu}{\nu_0}\frac{\delta(\nu_0)}{\nu_0}\right]^2}, \tag{4}$$

where $\delta(\Delta\nu)$ and $\delta(\nu_0)$ are the uncertainties in the shift in the beat frequency and in the empty cavity frequency, respectively.

The first term under the square root sign, which comprises the $\frac{\delta(\Delta\nu)}{\nu_0}$ entity, provides the contribution from the uncertainty in the assessment of the shift in the beat frequency when the measurement cavity is being filled with (or evacuated of) gas [29] . The second term, i.e. the $\frac{\Delta\nu}{\nu_0}\frac{\delta(\nu_0)}{\nu_0}$, depends on the uncertainty of the empty cavity frequency at the time instant of the refractivity assessment, i.e. $\delta(\nu_0)$. Since the latter term is proportional to the shift in the beat frequency, i.e. $\Delta\nu$, while the former is not, the former represents the uncertainty in refractivity that prevails under vacuum conditions (since, for an empty cavity, $\Delta\nu = 0$). The second term thus provides the additional contribution to the uncertainty in refractivity that appears due to the gas that has been filled into the cavity.

It should be noted though that the second term does not always contribute to the uncertainty when gas is present; it does not do so whenever the shift in the beat frequency is zero. Since this entity is reset to zero for each mode hop (which are made to accommodate large shifts of the cavity mode frequencies as gas is let into the cavity) [30], which takes place each time the shift in frequency reaches the FSR of the cavity, denoted ν_{FSR}, the uncertainty in the refractivity has a saw-tooth-like shape as a function of refractivity, ranging from $\frac{\delta(\Delta\nu)}{\nu_0}$ up to a maximum value of $\left(\left[\frac{\delta(\Delta\nu)}{\nu_0}\right]^2 + \left[\frac{\nu_{FSR}}{\nu_0}\frac{\delta(\nu_0)}{\nu_0}\right]^2\right)^{1/2}$.

Noting that the $\frac{\nu_0}{\nu_{FSR}}$ entity is a good representative of the number of the mode that is addressed by the laser, i.e. m_0, this implies that, to certify that a refractometry system based on a dual frequency measurement methodology can assess refractivity under low refractivity conditions with an uncertainty below a given benchmark, $\delta(n-1)_{BM}$, the expression

$$\sqrt{\left[\frac{\delta(\Delta\nu)}{\nu_0}\right]^2 + \left[\frac{1}{m_0}\frac{\delta(\nu_0)}{\nu_0}\right]^2} < \delta(n-1)_{BM} \tag{5}$$

needs to hold.

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3.2.2. Contributions to the uncertainty in the assessed refractivity – Limiting conditions

In general, the two uncertainties defined above, i.e. $\delta(\Delta\nu)$ and $\delta(\nu_0)$, have contributions from a number of sources that can (and will) affect the ability to assess refractivity. As is discussed in some detail in Section 1.2 in the Supplement 1, they originate predominantly from processes and entities such as:

(i) the assessments of the shift in the beat frequency and the empty cavity laser frequency [denoted $\delta(\Delta\nu)_{as}$ and $\delta(\nu_0)_{as}$], respectively;

(ii) the stability of the locking of the lasers to the cavity modes [$\delta(\Delta\nu)_{lock}$ and $\delta(\nu_0)_{lock}$];

(iii) the residual amount of gas in the cavity when being evacuated [$\delta(\Delta\nu)_{res}$ and $\delta(\nu_0)_{res}$]; and

(iv) the alterations in the length of the cavity that take place during the time intervals between two assessments [denoted $\delta(\Delta\nu)_{L_0}$ and $\delta(\nu_0)_{L_0}$, respectively].

As is shown by Eq. (S2) in the Supplement 1, this implies that Eq. (5), under the square root, in reality comprises eight squared terms.

The same equation shows that a necessary condition for the various $\delta(\Delta\nu)_i$ terms is that none of them should exceed $\nu_0\delta(n-1)_{BM}$, which henceforth will be referred to as the frequency difference condition. The same equation also shows that none of the $\delta(\nu)_i$ terms should exceed $\nu_0 m_0\delta(n-1)_{BM}$, which will be denoted the optical frequency condition. Inserting the pertinent values for the system scrutinized in this work (i.e. a refractivity benchmark of 2.7×10^{-12}, a wavelength of 1.55 μm, and an m_0 close to 2×10^5) implies that the frequency difference condition demands that none of the $\delta(\Delta\nu)_i$ terms should exceed 540 Hz while the optical frequency condition is significantly more relaxed; it requires that none of the $\delta(\nu_0)_i$ terms should exceed 110 MHz.

As is discussed in some detail in Section 1.3 in the Supplement 1, several of the aforementioned processes and entities [given by the points (*i*) to (*iv*) above], can, in a well-characterized system, be assessed with such high accuracy they do not violate any of the aforementioned conditions (which implies that they do not significantly contribute to the uncertainty of the refractivity at the level of the benchmark). This is in particular the case with the stability of the locking of the lasers to the cavity modes, which contributes to the $\delta(\Delta\nu)_{lock}$ and $\delta(\nu_0)_{lock}$ uncertainties in point (*ii*), the residual amount of gas in the cavity when evacuated, which gives rise to the $\delta(\Delta\nu)_{res}$ and $\delta(\nu_0)_{res}$ uncertainties in point (*iii*), and the assessment of the shift in the beat frequency, which contributes to the first uncertainty in point in (*i*), i.e. $\delta(\Delta\nu)_{as}$. This implies that Eq. (5) can predominantly be written solely in terms of three entities, viz. the relative uncertainties that are caused by the drifts in the length of the cavity, i.e. $\frac{\delta(\Delta\nu)_{L_0}}{\nu_0}$ and $\frac{\delta(\nu_0)_{L_0}}{\nu_0}$, given by the two terms in point (*iv*), and the one that is caused by the assessed empty cavity laser frequency, i.e. $\frac{\delta(\nu_0)_{as}}{\nu_0}$, i.e. the second term in point (*i*), as

$$\sqrt{\left[\frac{\delta(\Delta\nu)_{L_0}}{\nu_0}\right]^2+\left(\frac{1}{m_0}\right)^2\left\{\left[\frac{\delta(\nu_0)_{L_0}}{\nu_0}\right]^2+\left[\frac{\delta(\nu_0)_{as}}{\nu_0}\right]^2\right\}}<\delta(n-1)_{BM}. \tag{6}$$

As can be concluded from this equation, to fulfill the benchmark condition, also each of these three terms under the square root sign must individually be smaller that the benchmark.

The first term, i.e. $\frac{\delta(\Delta\nu)_{L_0}}{\nu_0}$, comprises the uncertainty in the shift in the beat frequency that is caused by the drift in length of the cavity between the instants of the measurements with and without gas in the measurement cavity, respectively. This shows that, to reach the benchmark, $\delta(\Delta\nu)_{L_0}$ cannot exceed the frequency difference condition, i.e. $\nu_0\delta(n-1)_{BM}$ (which in our case is 540 Hz). In addition to use a system that is well temperature-stabilized, as is shown in Section

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2 of the Supplement 1, this term can be held low by utilizing a short time separation between the two measurements and an interpolation procedure to assess the empty cavity mode frequency at the time instant when the refractivity is assessed.

The second term, which comprises $\frac{\delta(\nu_0)_{L_0}}{m_0\nu_0}$, is affected by the uncertainty in the empty cavity laser frequency that is caused by the drift in the length of the cavity between the instants of calibration of the laser frequency (i.e. the assessment of ν_0) and the measurement of the shift in the beat frequency, i.e. $\Delta\nu$. Since the time between these instants can be significant, the drifts can have a multitude of causes, comprising (but not being limited to) aging and temperature drifts. One should note that, in this term, since it has a $1/m_0$ dependence, the requirement of the drift in the frequency of the laser light on length, is significantly less strict than the corresponding requirement on the assessment of the shift in the beat frequency given by the first term. To reach the benchmark, it is, in fact, sufficient that $\delta(\nu_0)_{L_0}$ does not exceed the optical frequency condition, $m_0\nu_0\delta(n-1)_{BM}$ (which in our case is 110 MHz) [31]. It thereby suffices, in general, to utilize a well temperature-stabilized system with limited amounts of aging to hold this term below the benchmark. Alternatively, this term can be reduced by assessing ν_0 on a regular basis, e.g. by using the method described in this work.

The third term, which contains $\frac{\delta(\nu_0)_{as}}{m_0\nu_0}$, represents the uncertainty contribution from the assessment of ν_0. To reach the benchmark condition, this term has the same requirement on the laser frequency as the second one above, i.e., that the uncertainty of the assessment of the laser frequency needs to be below the optical frequency condition, in our case 110 MHz.

This shows that, in order to reach the benchmark condition, under the condition that the two length drift terms above can be held below their benchmark conditions, which, as is mediated above, is fully possible with a proper set of procedures, it is necessary to find means to assess ν_0 with an uncertainty below the optical frequency condition, preferably also under conditions when no laser frequency measuring instrumentation is available. As was alluded to above, this work presents a procedure that can achieve this.

4. Procedure for low uncertainty assessment of empty cavity mode frequencies in FPC based refractometry system without the use of laser frequency measuring instrumentation

The procedure for low uncertainty assessment of empty cavity mode frequencies without access to laser frequency assessment instrumentation requires that the cavity addressed previously has been characterized with respect to its physical parameters, viz. the phase shift of the light at the mirrors upon reflection and the Gouy phase (i.e. in terms of γ_c and Θ_G, respectively) and that the number of the mode addressed under vacuum conditions (m_0) has been unambiguously assessed. A method for how to assess these entities is described in Silander et al. [23,32,33].

Since it has become commonplace to utilize dual FPCs (DFPCs) in FP-based refractometry, and a description for assessments of refractivity in such have been worked out [23], it is suitable to describe the procedure for the case when a DFPC is used. The procedure used is based on measuring the beat frequency between the two cavities (denoted "1" and "2", representing the measurement and the reference cavity, respectively) under vacuum conditions while a forced mode jump is made in the cavity that is under investigation (here assumed to be cavity "1"). It has been found convenient to base the procedure on the "unwrapped" beat frequency, f_{UW}, (described in Silander et al. [12]), by which the wrapped beat frequency, f, is "corrected" for any possible mode jumps, given by

$$f_{UW} = \pm f - \left(\frac{\Delta m_1}{m_{01}}\nu'_{01} - \frac{\Delta m_2}{m_{02}}\nu'_{02}\right), \tag{7}$$

where Δm_1 and Δm_2 are the shifts of the numbers of the modes addressed when f_{UW} is assessed from those for which the system once was characterized (denoted m_{01} and m_{02}, respectively),

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defined as $m_1 - m_{01}$ and $m_2 - m_{02}$, where m_1 and m_2 are the mode numbers at which f_{UW} is assessed, respectively. The ν'_{01} and ν'_{02} entities represent the phase-shift-and-Gouy-phase-corrected empty cavity laser frequencies for the measurement and reference cavities defined as, for i = 1 or 2, $\nu_{0i}/\left(1 + \frac{\Theta_{G,i}}{\pi m_{0i}} + \frac{\gamma_{c,i}}{m_{0i}}\right)$, respectively, and where the ± sign corresponds to the cases when the empty cavity laser frequency of the measurement cavity is higher and lower than that of the reference cavity, respectively.

By use of this concept , and the condition that, when the measurement cavity is evacuated, this entity should be continuous during mode jumps, i.e. that the assessed refractivity should not change when a mode jump takes place, it is possible to derive expressions for the empty cavity laser frequencies of both the measurement and reference cavities in terms of previously characterized system parameters and measurable entities.

For the case when the laser addressing the measurement cavity is forced to make a mode jump (for simplicity assumed to take place from the mode at which the system was characterized, i.e. for $\Delta m_1 = 0$, to a neighboring mode, i.e. for $\Delta m_1 = 1$), while the laser addressing the reference cavity stays on the same mode (for simplicity assumed to be the one on which that cavity was characterized i.e. for $\Delta m_2 = 0$), it is possible, from the requirement that the "unwrapped" beat frequency should be continuous, i.e. that $f_{UW}(\Delta m_1 = 0) = f_{UW}(\Delta m_1 = 1)$, to conclude that

$$\pm f(\Delta m_1 = 0) = \pm f(\Delta m_1 = 1) - \frac{1}{m_{01}} \nu'_{01}. \tag{8}$$

By noting that the absolute value of the difference between these two beat frequencies, i.e. $|f(\Delta m_1 = 1) - f(\Delta m_1 = 0)|$, for future use denoted $(\Delta f)_1$, is equal to the FSR of the cavity, $\nu_{FSR,1}$, it is possible to conclude that this expression can be written as

$$\nu_{FSR,1} = \frac{\nu'_{01}}{m_{01}}. \tag{9}$$

This shows that, by performing as assessment of the FSR by the use of induced mode jumps, and by use of the definition of the phase-shift-and-Gouy-phase-corrected empty cavity laser frequency of the cavity addressed (i.e. ν'_{01}, as defined above), it is possible to assess the empty cavity laser frequency of the measurement cavity, ν_{01}, from an assessment of the FSR according to [34]

$$\nu_{01} = \nu_{FSR,1} \left(m_{01} + \frac{\Theta_{G1}}{\pi} + \gamma_{s,1} \right). \tag{10}$$

The empty cavity laser frequency of the reference cavity can be assessed by a corresponding expression in which all subscripts of "1" is exchanged to "2".

5. Experimental

The experimental setup used in this work, which is shown in Fig. 1, is virtually identical to that used in Silander et al. [23], to where the reader is referred for a more in-depth technical description of the system. In short it comprises the following parts.

The cavity system used is the same Invar-based DFPC as was used in some previous works [35–37]. The lasers used were Er-doped fiber lasers emitting light at around 1.55 μm. The mirrors were produced by a major producer of mirrors, with a refractivity of 99.97% at a center wavelength of 1.525 µm.

The lasers were locked to the cavity modes by the Pound-Drever-Hall (PDH) locking scheme. The system has incorporated a relocking routine that automatically (within a tenth of a second) performs a controlled jump of the frequency of the laser to a neighbouring cavity mode as soon as its frequency is outside a given preset range. This means that the utilized scanning ranges of the lasers are in the order of the FSR of the cavities, which are close to 1 GHz (see below).

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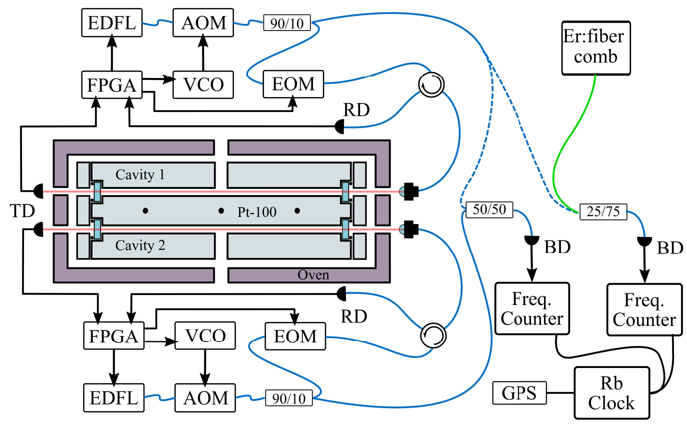

Fig. 1. Schematic illustration of the experimental setup. EDFL (Er-doped fiber laser), AOM (acousto-optic modulator), 90/10 (fiber splitter), EOM (electro-optic modulator), RD (fast photodetector for the reflected light), TD (large area photodetector for the transmitted light), FPGA (field programmable gate array), VCO (voltage-controlled oscillator), 50/50 (fiber coupler), BD (fast fiber-coupled photodetector for the beat signal), TD (transmission detector), Freq. Counter (frequency counter), Er:fiber comb (Er fiber-based frequency comb), and 25/75 (fiber coupler). Black arrows represent electrical signals, blue and green curves represent optical fibers, and red solid lines free-space beam paths. The dashed blue curves represent two different modes of operation; 1) when the FSR was assessed, the two fibers from the two lasers were sent to the 50/50 beam coupler or 2) when the laser frequency was assessed, the laser, here exemplified by the upper EDFL, was sent to a 25/75 beam coupler where it was combined with the frequency comb. Reproduced with permission from [23].

The mode jump routine is automatically engaged when the shift of the frequency of the laser probing the measurement cavity exceed a FSR but can also be user-initiated when the FSR of any of the cavities is to be assessed. To mitigate the influence of disturbances, and to make the present study feasible, the GAMOR methodology, which has a number of appealing properties and abilities, was used [12,23,37–44].

Specifically in this work, to measure the frequencies of the locked lasers, they were sequentially beat with the light from an Er:fiber frequency comb (Menlo Systems, FC1500-250-WG), referenced to a GPS-disciplined Rb clock with a relative accuracy of 5×10^{-12} over 1 s. This was done by merging the light from the locked laser under scrutiny [the blue dashed curves in Fig. (1)] with the light from the frequency comb (the green curve) by use of a 25/75 beam combiner that merged the two laser fields and sent them onto a beat detector (BD) whose beat signal was detected by a separate frequency counter referenced to the Rb clock.

6. Results

The empty cavity laser frequencies of both cavities in the DFPC system were assessed by the procedure presented. To verify the results, they were thereafter assessed also by the use of a frequency comb.

6.1. Assessment of empty cavity laser frequencies by use of the presented procedure

The FSR:s of the two cavities, $\nu_{FSR,1}$ and $\nu_{FSR,2}$, were repeatedly measured, as described around Eq. (10) above, by detecting the changes in the beat frequencies, $(\Delta f)_1$ and $(\Delta f)_2$, respectively, while changing the mode number of the cavity addressed by one unit (performed by repeatedly unlocking the laser and relocking it to an adjacent cavity mode).

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Figure (2) displays, in panel (a), the beat frequency from such a scrutiny of cavity 1, denoted f_1, with its associated shift when the number of the cavity mode addressed in the measurement cavity is changed by one unit (at 15 and 30 s, respectively). Panel (b) displays, by the colored curves, the change in beat frequency, $(\Delta f)_1$, from ten consecutive such measurements of the same cavity, while the solid black curve illustrates their average.

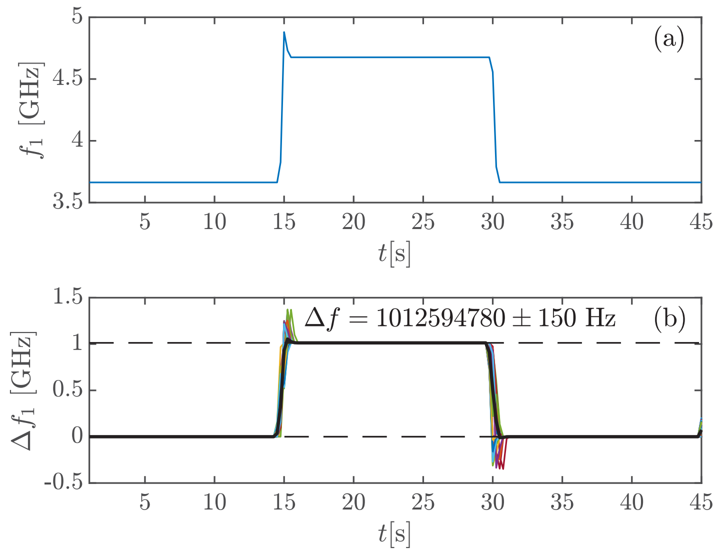

Fig. 2. Assessments of the beat frequency and the FSR from the measurement cavity. Panel (a) displays the beat frequency for cavity 1, i.e. f_{01} (representing ν_{01}), for a single FSR measurement with forced mode jumps taking place at 15 and 30 s, respectively, while panel (b) shows, by the set of colored curves, the change in the same entity, i.e. $(\Delta f)_1$, for ten consecutive measurements, and, by the solid black curve, their average.

Based on this, the FSRs of the two cavities could be assessed (with $k = 2$ uncertainties) to 1.012 594 78(15) and 1.012 586 41(15) GHz for the measurement and reference cavities, respectively, taken over a time corresponding to the optimum measurement time given by an Allan plot, i.e., for a few minutes and longer [45].

By use of Eq. (10), the previously assessed values of the phase shifts of the light at the front facets of the mirrors and the Gouy phases for the cavities [i.e. the $\gamma_{s,i}$ and $\frac{\Theta_{G,i}}{\pi}$, respectively, which, for both cavities, were assessed to 1.728(32) and 0.253(2)], and the assessments of the FSRs of the two cavities, the empty cavity laser frequencies of the measurement and reference cavities could be assessed to 193 401.53(3) and 193 397.91(3) GHz, respectively. This shows that the empty cavity laser frequencies could be assessed with an uncertainty ($k = 2$) of 30 MHz, which corresponds to a relative uncertainty of 0.15 ppm.

Note that the assessments of the shift of the mode frequency were made with the frequency counter referenced to the GPS-disciplined rubidium clock. Since the precision of the frequency counter was better than the uncertainty in the assessed FSR (150 Hz), this is though not required.

6.2. Verification of the assessed empty cavity laser frequencies by the use of a frequency comb

To verify the empty cavity laser frequencies assessed by the presented procedure, they were also assessed by the use of a frequency comb. The same method as was outlined in Silander et al. [23] was used, which, according to Eq. (21) in that work, assesses the frequency of the laser light, f_{0i} (denoted f_{cw} in [23]), as the sum of (or difference between) the frequency of one teeth of the frequency comb and the beat frequency between the frequencies of the laser and the closest frequency comb tooth (denoted f_b). Since, for such assessments, the laser investigated is locked to a mode of the cavity, f_{0i} represents, for laser i, ν_{0i}.

As in Silander et al. [23], it was found convenient to visualize the data in terms of plots of the difference in the assessed laser frequencies between two such measurements performed with dissimilar repetition rates of the frequency comb, denoted $f_{0i,m}$ and $f_{0i,n}$, respectively, as a function of comb tooth number (where the latter, for simplicity, is expressed as Δk, defined as the difference to an estimated comb tooth number).

Figure (3) shows, by the panels (a) and (b), this entity for six sets of measurements addressing the measurement and the reference cavities, respectively, where the various sets of data represent, for the measurement cavity, the differences $f_{0i,1} - f_{0i,6}$ to $f_{0i,6} - f_{0i,6}$ and, for the reference cavity, the differences $f_{0i,1} - f_{0i,1}$ to $f_{0i,6} - f_{0i,1}$, as a function of Δk. The data in each panel of the figure show that the various pairs of assessments produce the same laser frequency (i.e., a $\Delta f_{0i} = 0$) for a Δk value of zero. This confirms that the initial estimate of the tooth numbers was correct.

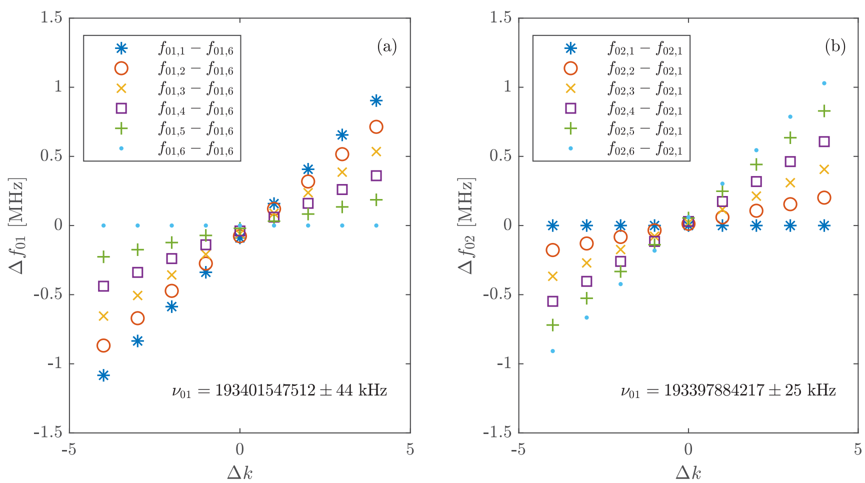

Fig. 3. Confirmation that the assumed tooth numbers was correct. Panels (a) and (b): the difference in the assessed laser frequencies between two measurements performed with dissimilar repetition rates of the frequency comb as a function of the estimated comb mode number, Δk, for the case with the laser locked to the measurement and reference cavity, respectively. The six sets of curves in panel (a) represent $f_{01,1} - f_{01,6}$ to $f_{01,6} - f_{01,6}$ while those in panel (b) correspond to $f_{02,1} - f_{02,1}$ to $f_{02,6} - f_{02,1}$, respectively. The correct comb tooth number is given by the one for which all predicted laser frequency differences are the same, which takes place for $\Delta k = 0$. Based on this, together with knowledge about the repetition rate and the frequency offset of the comb, the frequencies of the two lasers locked to the measurement and reference cavities could be assessed to 193 401.547 512(44) and 193 397.884 217(25) GHz, respectively

Based on knowledge about the repetition rate and the frequency offset of the comb, this implies that the frequencies of the two lasers could be assessed to 193 401.547 512(44) and 193 397.884 217(25) GHz, respectively. Since these assessments could provide frequency measurements with significantly smaller uncertainties than those by the procedure presented in this work, viz. 44 and 25 kHz ($k = 2$) [46] and, as is shown by the side-by-side comparison given in Table 1, since the frequencies are well within the uncertainties of the assessments made by the procedure presented above, they verify that the empty cavity laser frequencies assessed by the procedure presented in this work agree well with the actual laser frequencies.

Table 1. Comparison between the empty cavity frequencies of the two cavities assessed by the method presented in this work and the frequency comb.

	Cavity 1 (GHz)	Cavity 2 (GHz)
The presented method	193 401.53(3)	193 397.90(3)
The frequency comb	193 401.547 512(44)	193 397.884 217(25)

7. Summary and conclusions

This paper first shows that when a well-characterized FPC [47] is used for assessment of refractivity in the low refractivity regime, defined as the refractivities that are mainly limited by refractivity-independent sources of uncertainty, there are three main contributions to the uncertainty in the assessed refractivity, given by the three separate terms under the square root sign in Eq. (6) . These represent, in turn, (*i*) the uncertainty in the assessment of the shift in the beat frequency due to the alterations of the length of the cavity that take place during the time interval between the assessments with and without gas in the measurement cavity, respectively, (*ii*) the uncertainty in the assessment of the laser frequency due to the changes of the length of the cavity that take place between the assessment of ν_0 and the measurement of the shift in the beat frequency, and (*iii*) the uncertainty in the assessment of the empty cavity laser frequency, respectively.

While the two length drift terms both can be held below the benchmark condition (corresponding to an uncertainty of 1 mPa when nitrogen is addressed) by the use of appropriate measurement procedures [48], the uncertainty contribution from the assessment of ν_0 needs to be assessed by some experimental means with sufficiently low uncertainty.

It is thereafter shown that under the conditions that the appropriate measurement procedures have been addressed adequately, a requirement on the uncertainty in the assessment of the empty cavity laser frequency, i.e. on $\delta(\nu_0)_{as}$, to reach, under low refractivity conditions, a refractivity-independent benchmark, $\delta(n-1)_{BM}$, of 2.7×10^{-12}, which corresponds to an uncertainty in assessment of nitrogen at room temperature of 1 mPa, is given by an entity referred to as the optical frequency condition, given by $m_0 \nu_0 \delta(n-1)_{BM}$. For the case with a laser frequency of 1.55 μm, and when the number of the mode addressed is around 2×10^5, this corresponds to an uncertainty in the assessment of the empty cavity laser frequency, $\delta(\nu_0)_{as}$, that is 110 MHz [49]. It is therefore concluded that it is of high importance to find means to assess ν_0 with an uncertainty below this value.

Although there are techniques to assess the frequency of laser light with this uncertainty, such types of instrumentation might not always be available at the time of the assessment of refractivity (which often is the case when refractivity is assessed by the use of transportable refractometry systems), or, if being available, they can require complex and time consuming measurement procedures (e.g., when a frequency comb is used). The uncertainty in the assessment of the empty cavity frequency can then become the limiting factor in the assessment of refractivity by use of FPC-refractometry.

As a remedy to this, this paper has presented, in section 4, a procedure for how to assess the frequency of a laser addressing a mode in a well-characterized FPC-system that can, without access to any laser frequency measuring instrumentation (e.g., a wavelength meter or a frequency comb) at the time of the assessment of the refractivity, provide assessments of the laser frequency with an uncertainty that is well below the condition given above. The procedure is based on the fact that the assessed refractivity needs to be continuous (i.e. that is should not change) when mode jumps take place. It requires a well-characterized system, i.e. a system whose phase shift of the light upon reflection by the mirrors and Gouy phase are known, and knowledge about the number of the mode that the laser addresses (which can have been assessed as a part of a previous characterization) [50], and makes use of an assessment of the FSR of the cavity, i.e. the shift in frequency that takes place when a mode jump is induced.

It is shown that the empty cavity laser frequencies of the measurement and reference cavities addressed in this work could be assessed to 193 401.53(3) and 193 397.90(3) GHz, respectively, thus with (k = 2) uncertainties of 30 MHz, which corresponds to an uncertainty in pressure for nitrogen of 0.3 mPa, well below the benchmarks of 110 MHz and 1 mPa.

To verify the assessed laser frequencies, simultaneous assessments were made by use of a frequency comb. As is shown by Table 1, the assessments indicated that the frequencies of the two lasers, which were assessed to 193 401.547 512(44) and 193 397.884 217(25) GHz, respectively, were well within the uncertainties of the assessments by the presented method. This confirms the validity of the method presented.

The presented procedure for assessment of the empty cavity frequencies, which, as is described in Section 6.1 above, comprises assessments of a number of FSR:s of the two cavities while actively changing the mode number of the cavity addressed by one unit, can be performed within one hour, which makes it possible to repeatedly, within a measurement campaign, calibrate the empty cavity laser frequency. This implies that, once the FPC system has been characterized with respect to its physical properties and the number of the mode the laser addresses under vacuum conditions has been assessed, assessments of refractivity down to the benchmark condition can, under low refractivity conditions, be performed without access to any laser frequency measuring instrumentation. This makes the system self calibrating with respect to the laser frequency and the procedure can also be automated.

The presented method is not only expected to be useful to reach the $\delta(n-1)_{BM}$ benchmark (which represents the refractivity that corresponds to a pressure of nitrogen of 1 mPa) under normal laboratory conditions when no laser frequency measuring instrumentation with sufficient accuracy (or no such instrumentation at all) is available, it is particularly useful when transportable refractometers are being used or when calibrations or pressure assessments are to be made in industrial environments.

In addition, in cavities that have large drifts, e.g. due to aging (like in glass cavities), assessments of ν_0 and its drift during measurements are crucial. By regularly measuring ν_0 during a measurement campaign by the procedure presented in this work, its drift can be monitored accurately and repeatedly during a measurement campaign which, if needed, can provide means to reduce the uncertainty from elusive drifts in the length of the cavity.

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Data availability. Data underlying the results presented in this paper may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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13. When pressure is assessed, the assessment is also affected by the molecular constants (the molar polarizability and the relevant virial coefficients) of the gas addressed and the assessment of the gas temperature. However, since the present study deals with a procedure for automated and repeated assessments of the laser frequency in FPC-based refractometers, which affects the ability to assess refractivity, the concepts of molecular constants and assessment of the gas temperature are not addressed in this work.
14. Cavity drifts can be compensated for either by the use of estimates based on the long term behavior of the cavity (e.g. by the use of extrapolations of previously performed assessments of cavity lengths), by the use of a measurement procedure that eliminates (or reduces significantly) the influence of such drifts on the measurement process (as is used in the Gas Modulation Refractometry (GAMOR) methodology [12,35–44]), or by repeated assessments of the empty cavity mode frequency, e.g. by the procedure that is presented in this work.
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21. The assessment of the number of the mode addressed can have been carried out at an earlier performed characterization which, at that time, have required access to laser frequency measuring instrumentation with an uncertainty that does not exceed half of the free-spectral-range of the cavity [23].
22. It is also shown that the use of a dual frequency measurement methodology with short time separations between frequency assessments with and without gas in the cavity and where the latter ones are assessed by the use of interpolation, as is used in the GAMOR methodology [12,35–44], can reduce the influence of several types of drifts, in particular those from the length of the cavity.
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27. As is shown below, the fact that the assessment of $\Delta\nu$, often is substantially separated (time-wise) from the assessment of the frequency of the laser light, ν_0 (which henceforth will be referred to as the "calibration of the laser frequency"), does not adversely affect the ability of the methodology to assess refractivity.
28. The characterization implies that the number of the mode the laser addresses under vacuum conditions, m_0, is known, and that the number of mode jumps that have taken place during filling (or emptying) of the cavity, Δm, has been assessed. It has been shown by Silander et al. [23] that these can be assessed with no uncertainty. The phase shift

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of the light at the front facets of the mirrors upon reflection and the Gouy phase can be assessed with such small uncertainties that they will not affect a system to a level comparable to that of the benchmark. It also follows, from the definition of the low refractivity regime above, that the assessment of refractivity in this regime is not affected by the uncertainty of the assessment of the distortion.
29. Since cavity mode frequencies are commonly assessed as beat frequencies with another field by the use of a frequency counter, the $\delta(\Delta\nu)$ entity represents, in practice, the uncertainty in the assessment of beat frequency assessed by a frequency counter.
30. The mode hops still contribute to the assessment of refractivity through the Δm term in Eq. (2).
31. This implies that when the dual frequency measurement methodology is used together with a well temperature-stabilized system, the time separation between the calibration of the empty cavity laser frequency and the assessment of refractivity when there is gas in the cavity can be substantial without adversely affecting the ability to assess refractivity with an uncertainty at the level of the benchmark.
32. As is described in some detail in Silander et al. [23], such an assessment commences with an assessment of the number of the mode addressed, m_0. To do so requires an ability to assess the laser frequency with an uncertainty that is below half a free-spectral-range of the FPC, i.e. $<\nu_{FSR}/2$, which in our case is 500 MHz, as well as an approximate estimate of γ_c. While the former can be assessed by the use of any wavelength meter that has an uncertainty that is better than $\nu_{FSR}/2$, the latter can be estimated by the use of a standard expression comprising the indices of refraction of the two types of layers that make up the QWS of the mirror coating. The Gouy phase can be assessed from the geometrical shape of the cavity (its length and the radii of the mirrors).
33. Since a well-stabilized low drift cavity can have a drift rate well below 10^{-6} year^{-1} (which corresponds to 3×10^{-14} s^{-1}), the system will remain at the mode number assessed by the characterization for a substantial amount of time, in the order of years. This implies that it is sufficient that the characterization has been done within such a time frame before the assessment of refractivity takes place, which, in tun, implies that looking for a change in the empty cavity mode number (e.g., by monitoring the laser piezo voltage) can be done quite irregularly.
34. Note that in Eq. [10], the $\gamma_{c,i}$ has been replaced with $\gamma_{s,i}$, which has to be used in the case when the laser frequency is not on the design frequency of the mirror [23].
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47. One whose physical properties, primarily the phase shifts of the light at the front facets of the mirrors upon reflection and the Gouy phase for the cavity, have been assessed with low uncertainties, and when the number of the cavity mode the laser addresses is known.
48. For example, by the use of a dual frequency measurement methodology together with the gas modulation refractometry (GAMOR) methodology [12,35–44] or the use of the procedure to assess ν_0 presented in this work.
49. This implies that the relative uncertainty in the assessment of the empty cavity laser frequency needs to be $m_0\delta(n-1)_{BM}$, which in our case is 0.5 ppm.
50. When a given cavity mode has been chosen as suitable for assessments of refractivity under low refractivity conditions, and its mode number has been uniquely assessed, due to the restricted drifts in length of the cavity spacer and the good reproducibility of modern lasers to produce a given laser frequency for a given set of input parameters, the same cavity mode can, for a considerable amount of time (in the order of years), unmistakably be addressed any time subsequent to the assessment of the mode number.