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Cite as: J. Vac. Sci. Technol. B **38**, 054202 (2020); <https://doi.org/10.1116/6.0000375>

Submitted: 03 June 2020 . Accepted: 01 September 2020 . Published Online: 24 September 2020

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Submitted: 3 June 2020 · Accepted: 1 September 2020 ·

Published Online: 24 September 2020



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ABSTRACT

A novel procedure for a robust assessment of cavity deformation in Fabry–Pérot (FP) refractometers is presented. It is based on scrutinizing the difference between two pressures: one assessed by the uncharacterized refractometer and the other provided by an external pressure reference system, at a series of set pressures for two gases with dissimilar refractivity (here, He and N₂). By fitting linear functions to these responses and extracting their slopes, it is possible to construct two physical entities of importance: one representing the cavity deformation and the other comprising a combination of the systematic errors of a multitude of physical entities, viz., those of the assessed temperature, the assessed or estimated penetration depth of the mirror, the molar polarizabilities, and the set pressure. This provides a robust assessment of cavity deformation with small amounts of uncertainties. A thorough mathematical description of the procedure is presented that serves as a basis for the evaluation of the basic properties and features of the procedure. The analysis indicates that the cavity deformation assessments are independent of systematic errors in both the reference pressure and the assessment of gas temperature and when the gas modulation refractometry methodology is used that they are insensitive to gas leakages and outgassing into the system. It also shows that when a high-precision (sub-ppm) refractometer is characterized according to the procedure, when high purity gases are used, the uncertainty in the deformation contributes to the uncertainty in the assessment of pressure of N₂ with solely a fraction (13%) of the uncertainty of its molar polarizability, presently to a level of a few ppm. This implies, in practice, that cavity deformation is no longer a limiting factor in FP-based refractometer assessments of pressure of N₂.

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I. INTRODUCTION

Refractometry constitutes an optical technique for the assessment of refractivity that, by the use of the Lorentz–Lorenz equation and an equation of state, can assess molar density and pressure with high precision and accuracy.^{1–4} It has been indicated that the technology also has the possibility to realize the Pascal.⁵ This opens up for a variety of usages of refractometry.

The most sensitive refractometers are based on Fabry–Pérot (FP) cavities in which a laser is used to probe the frequency of a longitudinal mode.^{6–13} Although different realizations of such refractometers have shown promising results, a crucial limiting issue that needs to be addressed is the fact that the cavities are subjected to pressure-induced deformation that will compress (or elongate) the cavities when they are exposed to gas pressure. Without taking this effect into consideration properly, pressure

assessments can be adversely affected on the permille to percent range.

As a means to reduce the effect of deformation, Egan *et al.*¹⁴ recently suggested (based on a work by Stone and Stejskal⁸) a procedure in which one gas, whose molar polarizability is known (He), is used to characterize the system with regard to its pressure-induced deformation, allowing for subsequent accurate assessments of the pressure of other gases. This is an ingenious idea that can facilitate the assessment of cavity deformation in FP-based refractometers. However, despite the fact that the suggested procedure does not require any knowledge about the pressure used for the deformation assessment (here referred to as the set pressure)—it is sufficient if it is constant—the way in which the deformation assessment is tacitly assumed to be done—by utilizing the difference between the responses when the two gases are addressed—potentially

opens up for influences of a number of physical processes, e.g., gas leakages and outgassing, that can reduce the accuracy of the assessed cavity deformation.

As a remedy to this, inspired by the work by Egan *et al.*,¹⁴ this paper presents a procedure for robust assessment of cavity deformation that is not only unaffected by any systematically incorrect assessment of the set pressure but also not influenced by a systematically incorrect assessment of the gas temperature. If the procedure is carried out by the use of the gas modulation refractometry (GAMOR) methodology,^{12,13,15} it is also insensitive to gas leakages and outgassing into the system.¹⁶ It is performed by assessing the difference between the pressure assessed by the uncharacterized refractometer and a supplied reference pressure at a series of pressures for two gases with dissimilar refractivity (here He and N₂). By then fitting linear functions to these responses, their slopes are used as a basis for the assessment of the pressure-induced deformation of the cavity. In addition to being insensitive to the aforementioned processes and entities, the procedure is also capable of providing an estimate of a combination of systematic errors of some system parameters (those of the temperature assessment, the estimated penetration depth of light into the mirrors, the molar polarizabilities, and the set pressure of the pressure reference).¹⁷

In this paper, we provide, predominately in theoretical terms, a thorough description of the procedure that not only explicates its principles but also provides a basis for evaluation of its basic properties and features. It is shown that the procedure, when carried out with a high-precision refractometer system, allows for an assessment of the deformation with an uncertainty in the low %-range (solely given in terms of the uncertainty of the molar polarizabilities of the gases addressed). This allows for assessments of the refractivity of the “second” gas in which the cavity deformation solely contributes to the uncertainty with a fraction of the uncertainty of its molar polarizability (for N₂ with 13% of the uncertainty of its molar polarizability). To exemplify its execution and applicability, we apply it to a cavity deformation assessment of a previously developed high-precision Invar-based refractometry system.¹⁸

II. THEORY—BASIC RELATIONS FOR ASSESSMENT OF PRESSURE BY REFRACTOMETRY

A. Refractivity

As is shown in some detail in Sec. I of the supplementary material,¹⁹ the refractivity for a gas i , $(n - 1)_i$, assessed by the use of a dual-Fabry–Pérot-cavity (DFPC) refractometer, can be expressed as a function of the shift of the beat frequency between two laser fields that each is locked to a mode in one of the two cavities, Δf , and, if mode jumps take place, the shift in mode number of the modes addressed in the measurement cavity, Δq , when the measurement cavity is evacuated (or filled with gas), alternatively as

$$(n - 1)_i = \frac{\Delta f + \Delta q}{1 - \Delta f} (1 + 2\overline{L}_{pd}) - \overline{\delta L} \quad (1a)$$

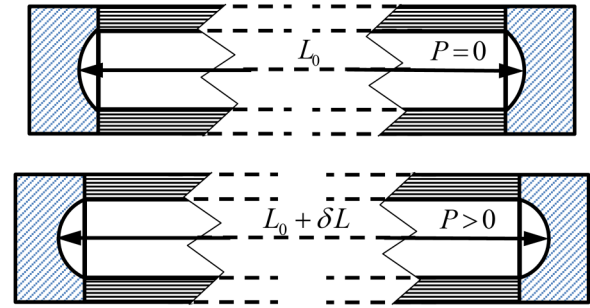


FIG. 1. Schematic illustration of the pressure-induced deformation of a single closed FPC exposed to a gas. For this type of cavity, the deformation will comprise both an elongation of the length of the cavity spacer and a bending of the mirrors. Note that δL is negative for an open cavity.

or as

$$(n - 1)_i = \frac{\overline{\Delta f} + \overline{\Delta q}}{1 - \overline{\Delta f} + \varepsilon_i} (1 + 2\overline{L}_{pd}), \quad (1b)$$

where $\overline{\Delta f}$ is given by $\Delta f/\nu_0$, where ν_0 is the frequency of the laser addressing the q_0 mode in the evacuated measurement cavity; $\overline{\Delta q}$ is given by $\Delta q/q_0$; \overline{L}_{pd} represents L_{pd}/L_0 , where L_{pd} is the light penetration depth in the mirrors, given by Eq. (SM-3) in the supplementary material,¹⁹ and L_0 is the empty measurement cavity length; and $\overline{\delta L}$ denotes $\delta L/L_0$, where δL , as is shown in Fig. 1, is the pressure-induced cavity deformation,²⁰ and where ε_i is a deformation parameter, defined as $\overline{\delta L}/(n - 1)_i$. These expressions are valid under the assumption that no mode jump occurs in the reference cavity. As has been mediated in previous publications,^{12,13} Eq. (1b) is also valid, with a minor redefinition of Δf , for the case when the measurement cavity is not fully evacuated but contains a minor residual gas pressure.

Under the condition that $\overline{\delta L}$ is linearly dependent on pressure, which is customary to assume, it can be written as κP . For a well constructed cavity, κ , which is an alternative deformation parameter, is typically in the order of 10^{-12} to 10^{-11} Pa⁻¹.²¹ It is shown in Sec. I of the supplementary material¹⁹ that Eq. (1a) has a relative accuracy of $\overline{\delta L}$ and Eq. (1b) a similar (or slightly better) relative accuracy ($\varepsilon_i \overline{\Delta q}$). This implies that, for pressures up to 10^5 Pa, they are accurate to at least 1 ppm.

There are a number of advantages of describing the cavity deformation in term of ε_i instead of $\overline{\delta L}$, predominantly that

- (1) it is, for a range of pressures (for conditions, see Sec. III of the supplementary material¹⁹), a constant, while $\overline{\delta L}$ is a function of pressure;
- (2) since $(n - 1)_i \propto (1 - \varepsilon_i)$, it represents, to first order, the relative influence of the cavity deformation to the assessed refractivity; and
- (3) its assessment by the novel procedure is not influenced by systematic errors in the gas temperature assessment.

B. Molar density

As is shown in the supplementary material,¹⁹ the molar density of a gas i , ρ_i , can be calculated from the refractivity, for gases without magnetizability, by the use of the extended Lorentz–Lorenz equation, and, for those with a non-negligible amount of magnetizability, by a pair of similar extended expressions for the relative dielectric and magnetic polarizabilities. In both cases, it can, for the range of pressures addressed in this work,²² be written as

$$\rho_i = \frac{2}{3A_{R,i}}(n-1)_i[1 + b_{n-1,i}(n-1)_i], \quad (2)$$

where $A_{R,i}$, for N_2 , is the dynamic molar polarizability, denoted A_{R,N_2} , while, for He, it is given by the sum of the dynamic molar dielectric and magnetic polarizabilities, given by $A_{\epsilon,He}$ and $A_{\mu,He}$, respectively. $b_{n-1,i}$ is a series expansion coefficient, whose leading term is $-(1 + 4b_{R,i}/A_{R,i})/6$, where $b_{R,i}$ is the second relative refractivity virial coefficient.^{1,13,23} For the case with a gas without magnetizability, the expression given by $b_{n-1,i}$ is exact. For gases with magnetizability, $b_{R,i}$ is the second relative dielectric refractivity virial coefficient, commonly referred to as $b_{\epsilon,i}$.²³

C. Pressure

The corresponding pressure, P , can then, again for the range of pressures addressed in this work,²⁴ be obtained from the density from

$$P = RT\rho_i[1 + B_{\rho,i}(T)\rho_i], \quad (3)$$

where R is the (ideal) gas constant (which is given by a product of the Avogadro number and the Boltzmann constant), T is the temperature of the gas sample, and $B_{\rho,i}(T)$ is the second pressure virial coefficient.

This implies that it is possible, for the range of pressures addressed in this work, to express the pressure in terms of the assessed refractivity according to¹⁹

$$P = \frac{2RT}{3A_{R,i}}[1 + \xi_{2,i}(T)(n-1)_i](n-1)_i, \quad (4)$$

where $\xi_{2,i}(T)$ is a series expansion coefficient given by $b_{n-1,i} + 2B_{\rho,i}(T)/(3A_{R,i})$.

Table SM-1 in the supplementary material¹⁹ provides a compilation of various series expansion coefficients for the expressions for density and pressure. Values from the literature for the leading virial coefficients [$A_{R,i}$, $B_{R,i}$, and $B_{\rho,i}(T)$] for N_2 and He, a pair of combinations of molar polarizabilities ($A_{R,He/N_2}$, which is defined as $A_{R,He}/A_{R,N_2}$ and $A_{R,N_2}/A_{R,He}$), and the (ideal) gas constant (R) are given in Table I, while Table SM-2 in the supplementary material¹⁹ summarizes relevant values for the leading series expansion coefficients [primarily $b_{n-1,i}$ and $\xi_{2,i}(T)$] and some common expressions containing such coefficients for N_2 and He. Note that Table I provides two slightly dissimilar values for the molar polarizability of nitrogen. They correspond to the best assessments that are traceable to a thermodynamic and mechanical standard, respectively.

TABLE I. Virial coefficients [$A_{R,i}$, $B_{R,i}$, and $B_{\rho,i}(T)$] for N_2 and He at 296.15 K and 1550.14 nm, a pair of combinations of molar polarizabilities, and the (ideal) gas constant. The uncertainties given refer to $k = 2$.

Parameter	Values	Reference
A_{R,N_2} ^a	$4.396\,546(138) \times 10^{-6} \text{ m}^3/\text{mol}^b$	14
	$4.396\,549(34) \times 10^{-6} \text{ m}^3/\text{mol}^c$	4 and 11
b_{R,N_2} ^d	$0.184(46) \times 10^{-6} \text{ m}^3/\text{mol}$	11
$B_{\rho,N_2}(T)^e$	$-5.34(24) \times 10^{-6} \text{ m}^3/\text{mol}$	11
$A_{\epsilon,He}$ ^a	$0.517\,753\,036(5) \times 10^{-6} \text{ m}^3/\text{mol}$	23
$A_{\mu,He}$	$-7.922\,4(4) \times 10^{-12} \text{ m}^3/\text{mol}$	23
$A_{R,He}$ ^a	$0.517\,745\,114(5) \times 10^{-6} \text{ m}^3/\text{mol}$	
$b_{\epsilon,He}$	$-0.104\,9 \times 10^{-6} \text{ m}^3/\text{mol}$	25
$B_{\rho,He}(T)^e$	$11.835(1) \times 10^{-6} \text{ m}^3/\text{mol}$	8
$A_{R,He/N_2}$	$0.117\,761\,8(37)^b$	
	$0.117\,761\,7(9)^c$	
$A_{R,N_2}/A_{R,He}$	$8.491\,72(27)^b$	
	$8.491\,72(7)^c$	
R	$8.314\,462\,6 \text{ J mol}^{-1} \text{ K}^{-1}$	26

^aAs is shown in Sec. IV of the supplementary material, the molar polarizabilities given here have been scaled from the temperature and wavelength at which they were assessed to the ones that are used in the present investigation (296.15 K and 1550.14 nm, respectively).

^bMolar polarizability of nitrogen at a wavelength of 1550.14 nm and a temperature of 296.15 K, and derived entities based on this, that are traceable to a thermodynamic pressure standard. The value given for A_{R,N_2} is based on a value $4.446\,107(138) \times 10^{-6} \text{ m}^3/\text{mol}$ assessed at a wavelength of 633 nm and a temperature of 293.15 K by Egan *et al.* (Ref. 14). The ($k = 2$) uncertainty of the A_{R,N_2} value given in the table corresponds to 31 ppm.

^cMolar polarizability of nitrogen at a wavelength of 1550.14 nm and a temperature of 296.15 K, and derived entities based on this, that are traceable to a mechanical pressure standard. The value given for A_{R,N_2} is based on a value $4.446\,139(30) \times 10^{-6} \text{ m}^3/\text{mol}$ assessed at a wavelength of 633 nm and a temperature of 302.919 by Egan *et al.* (Refs. 4 and 11). The ($k = 2$) uncertainty of the A_{R,N_2} value given in the table corresponds to 8 ppm.

^d b_{R,N_2} is given by $B_{R,N_2}/A_{R,N_2}$, where B_{R,N_2} is retrieved from Ref. 11.

^eAt a temperature of $T = 296.15(1) \text{ K}$.

D. Conclusive remarks—The advantage of assessing refractivity by use of Eqs. (1a) and (1b)

It is of importance to note that Eqs. (1a) and (1b) show that the refractivity (and thereby the molar density and pressure) can be assessed without any specific assessment or monitoring of L_0 . The reason for this is that the change in cavity mode frequency when the measurement cavity is evacuated (or filled with gas) is measured directly as a shift of a cavity mode, viz., as Δf .

There is neither any need to explicitly assess the free-spectral range (FSR). The reason for this is that Δq is not expressed in terms of the FSR but instead in the mode number of the mode addressed, q_0 , which is an integer that can be assessed uniquely (i.e., with no uncertainty) by ensuring that the assessed refractivity is a continuous function when the measurement laser is making a (controlled) mode jump.²⁷ It should be noticed that this does not require assessment of any physical entity (see Sec. VI of the supplementary material,¹⁹ for further discussion).

III. PROCEDURE

A. Assumptions and nomenclature

The procedure prescribes assessments of pressure by the use of the uncharacterized refractometer at a series of pressures. Although an appropriate assessment of pressure requires accurate knowledge about a number of physical entities (predominately, the molar polarizability of the gas addressed, the temperature of the gas, and the penetration depth), this is not needed for an accurate assessment of the cavity deformation by the presented procedure. To illustrate this, let us first assume that all of these are associated with errors. Let us also allow for that the system can be exposed to leakages and outgassing. The scrutiny of the procedure given below will then reveal to which extent the assessed deformation depends on these errors and processes.

Let us assume that the dynamic molar polarizabilities of the gases addressed, $A'_{R,i}$, are taken from the literature (for He, theoretically assessed and for N₂, experimentally assessed, both recalculated to the pertinent temperature and wavelengths) that systematically can differ from the actual (real or true) polarizabilities, $A_{R,i}$ s, by unknown amounts of $\delta A_{R,i}$ (defined as $A'_{R,i} = A_{R,i} + \delta A_{R,i}$). If needed, $\delta A_{R,i}$ can also represent any possible contamination in the supply of gas i . Hence, $\delta A_{R,i}$ represents the systematic error in $A'_{R,i}$.²⁸

Let us assume that the refractometry system considered has a stabilized cavity temperature that does not change markedly over the time over which the deformation assessment is performed. Let us further assume that the assessed temperature T' can systematically differ from the actual/correct temperature of the gas in the laser beams, T , by an amount of δT . The latter thus represents the systematic error in the assessment of temperature (defined by $T' = T + \delta T$). Let us assume that δT is independent of gas species and pressure.

In addition, let us assume that the estimated penetration depth, L'_{pd} , will differ from the actual/correct one, L_{pd} , by an amount δL_{pd} , which thus represents the systematic error in the estimation, defined as $L'_{pd} - L_{pd}$.

Moreover, let us also allow for the possibility that the system is exposed to (small) gas leakages or outgassing, with a constant leakage or outgassing rate of $(\partial P/\partial t)_{l/o}$.

Finally, let us allow for that the pressures set by the pressure reference can differ from the true ones with both a systematic (proportional) error of $\delta P_{Set,i}$, which can depend on the type of gas species used,²⁹ and a systematic (absolute) offset error δP_{Set}^0 .³⁰ This implies that the pressure reference system in reality provides a true pressure, $P_{True,i}$, that is given by $P_{Set} + \delta P_{Set,i} + \delta P_{Set}^0$.

B. Basic principles of the procedure

As is shown in Sec. V in the supplementary material,¹⁹ Eq. (4) indicates that, during the cavity deformation assessment process, the uncharacterized refractometer will assess the pressure supplied by the pressure reference system to be $P_{Refr,i}^U$ given by

$$P_{Refr,i}^U = \frac{2RT'}{3A'_{R,i}} [1 + \xi_{2,i}(T')(n-1)_i] (n-1)_i, \quad (5)$$

where $(n-1)_i$ is given by Eq. (1a) or (1b) with κ (and thereby $\overline{\delta L}$) and ε_i being zero.

This pressure differs slightly but noticeably from the true pressure ($P_{True,i}$), both because of the disregarded cavity deformation, κ or ε_i , and the systematic errors of the assessed temperature, δT , the molar polarizability, $\delta A_{R,i}$, the penetration depth, L_{pd} , and possible gas leakages and outgassing into the system. Note that it differs additionally from the set pressure, P_{Set} , by the systematic (constant and proportional) errors of the pressure reference, $\delta P_{Set,i}$ and δP_{Set}^0 . Note also that the only two pressures that are accessible to the experimentalist at this stage are $P_{Refr,i}^U$ and P_{Set} .

Since $P_{Refr,i}^U$ is similar to P_{Set} , it is often convenient to look at the difference between the two, denoted ΔP_i . It is shown by Eq. (SM-35) in Sec. V of the supplementary material¹⁹ that it is possible, for an ordinary well-designed system with small $\overline{\psi}_i$ and $\delta P_{l/o,i}$ (defined below), to express ΔP_i in terms of P_{Set} by the use of ε_i and $\overline{\psi}_i$ entities as

$$\begin{aligned} \Delta P_i &= \left(\varepsilon_i + \frac{\overline{\psi}_i}{1 - \overline{\psi}_i} \right) P_{Set} + \frac{\delta P_{l/o,i} + \delta P_{Set}^0}{1 - \overline{\psi}_i} \\ &\approx \left(\varepsilon_{0,i} + \frac{\overline{\psi}_i}{1 - \overline{\psi}_i} \right) P_{Set} + \delta P_{l/o,i} + \delta P_{Set}^0, \end{aligned} \quad (6)$$

where $\varepsilon_i = \varepsilon_{0,i} + \varepsilon_{B,i}$, in which $\varepsilon_{0,i}$, which is given by $\frac{2RT}{3A_{R,i}} \kappa$, is the leading term of ε_i , while $\varepsilon_{B,i}$ is a short hand notation for the influence of the nonlinear responses of the Lorentz-Lorenz equation and the equation of state on the cavity deformation, given by $\xi_{2,i}(T)(n-1)_i \varepsilon_{0,i}$, which alternatively can be written as $\xi_{2,i}(T) \kappa P_{Set}$. $\overline{\psi}_i$ represents a sum of the systematic errors of the aforementioned entities given by

$$\overline{\psi}_i = \overline{\delta T} - 2\overline{\delta L_{pd}} + \overline{\delta P_{Set,i}} - \overline{\delta A_{R,i}}, \quad (7)$$

where $\overline{\delta T}$, $\overline{\delta L_{pd}}$, $\overline{\delta P_{Set,i}}$, and $\overline{\delta A_{R,i}}$ represent $\delta T/T'$, $\delta L_{pd}/L'_{pd}$, $\delta P_{Set,i}/P_{Set}$, and $\delta A_{R,i}/A'_{R,i}$, respectively. $\delta P_{l/o,i}$, finally, referred to as the molar-polarizability-referenced gas leakage or outgassing pressure represents the influence of gas leakages or outgassing into the system, given by $P_{l/o} \Delta \overline{A_{R,l/o,i}}$, where $P_{l/o}$ is given by $(\partial P/\partial t)_{l/o} \Delta t$, where, in turn, Δt is the time since the last gas evacuation. $\Delta \overline{A_{R,l/o,i}}$ represents the relative difference between the molar polarizability of the leaking (or outgassing) gas, $A_{R,l/o}$, and that addressed, $A_{R,i}$, i.e. $(A_{R,l/o} - A_{R,i})/A_{R,i}$. Note that $\delta P_{l/o,i}$ is a pressure-independent entity that, for each gas species and for a constant gas leakage or outgassing process, is constant throughout the entire procedure. In addition, it becomes zero for the case when the leaking or outgassing gas has the same molar polarizability as the gas addressed.

It can be estimated, based on Sec. III in the supplementary material¹⁹ together with the assessed values for the cavity deformation (see below), that, for the pressure range used for the deformation assessment performed in this work, which is up to 16 kPa, the approximation made in the last step in Eq. (6) is considered adequate; it is smaller than half of the uncertainty that originates from the evaluation of the measurement data, which, in turn, is smaller than the relative systematic uncertainty in the molar polarizability

of N_2 , i.e., $\overline{\delta A'_{R,N_2}}$ (see below). Since $\varepsilon_{0,i}$ is a constant, this implies that ΔP_i can be considered to be linear with P_{Set} .

To provide an assessment of the cavity deformation with the aforementioned properties, ε_i and $\overline{\psi}_i$ are assessed by use of fits to data taken at several set pressures. This is done by assessing ΔP_i for a set of P_{Set} -values and fitting a first order function of the type $a_i + k_i \times P_{Set}$ to the data in the plots. A comparison with Eq. (6) implies that the linear coefficient of such a fit, k_i , can, for each gas, be related to the entities above as

$$k_i = \varepsilon_{0,i} + \frac{\overline{\psi}_i}{1 - \overline{\psi}_i}. \quad (8)$$

Note that the restriction in the pressure range used for the deformation assessment to that for which ΔP_i is linear with P_{Set} , i.e., the pressures for which ε_i can be approximated by $\varepsilon_{0,i}$, does not restrict the use of the characterized refractometer to the same pressure range.³¹

C. Expressions for assessment of pressure-induced cavity deformation, their systematic errors, and a combination of systematic errors of some system parameters

1. Pressure-induced cavity deformation

As is shown by Eq. (SM-38) in the supplementary material,¹⁹ eliminating in the two expressions for k_i in Eq. (8), viz., those for He and N_2 , the common $\delta T - 2\delta L_{pd}$ entity, provides an expression for κ in terms of the slopes for the two gases, the gas temperature, the molar polarizabilities of the gases addressed, and the difference in the systematic errors of the set pressure of the pressure reference system. However, since the temperature only can be experimentally assessed as T' and the molar polarizability is given as A'_{R,N_2} , the assessed value of κ , denoted κ' , will differ slightly from the “true” value of κ by an amount $\delta\kappa^s$, which represents its systematic error. As is shown by Eq. (SM-39) in the supplementary material,¹⁹ κ' and $\delta\kappa^s$ can be written as

$$\kappa' = \frac{3A'_{R,He}}{2RT'} \frac{k_{He} - k_{N_2}}{1 - A'_{R,He/N_2}}, \quad (9a)$$

$$\delta\kappa^s = \frac{3A'_{R,He}}{2RT'} \frac{(\overline{\delta A_{R,N_2}} - \overline{\delta A_{R,He}}) - (k_{He} - k_{N_2})\overline{\delta T}}{1 - A'_{R,He/N_2}}, \quad (9b)$$

where we have considered the pressure reference system to be species-independent, i.e., that $\delta P_{Set,He} = \delta P_{Set,N_2}$ and the influence of the penetration depth has been neglected with respect to the relative error in temperature. The case when this assumption does not hold is considered in the supplementary material.¹⁹

As also is shown in the supplementary material,¹⁹ based upon the definition of $\varepsilon_{0,i}$ and the expression for κ [Eqs. (SM-16) and (SM-38), respectively], it is possible to write similar expressions for the two experimentally assessed $\varepsilon'_{0,i}$'s and their systematic errors,

$\delta\varepsilon'_{0,i}$, as

$$\varepsilon'_{0,i} = \frac{A'_{R,He}}{A'_{R,i}} \frac{k_{He} - k_{N_2}}{1 - A'_{R,He/N_2}}, \quad (10a)$$

$$\delta\varepsilon'_{0,i} = \frac{A'_{R,He}}{A'_{R,i}} \frac{\overline{\delta A_{R,N_2}} - \overline{\delta A_{R,He}}}{1 - A'_{R,He/N_2}}. \quad (10b)$$

Equations (9a) and (10a) show that κ' and $\varepsilon'_{0,i}$'s take values that predominantly are given by the difference in slopes of the two ΔP_i vs P_{Set} plots, i.e., $k_{He} - k_{N_2}$, the molar polarizabilities of the two gases, i.e., $A'_{R,He}$ and A'_{R,N_2} , and (for κ') the assessed temperature, T' . They also show that the two deformation parameters are not affected by any virial coefficients (which is a consequence of the fact that the deformation assessment is performed at pressures for which the pressure-dependent deformation is linearly proportional to the refractivity—the conditions of which are discussed in Sec. III of the supplementary material¹⁹).

In agreement with the procedure suggested by Egan *et al.*¹⁴ and as is shown by Eqs. (9b) and (10b), the determinations of κ and $\varepsilon_{0,i}$ s are independent of the systematic error in the set pressure.

In addition to this, as is shown by Eq. (10b), the assessments of the $\varepsilon_{0,i}$ s are also independent of any systematic error in the assessment of gas temperature. κ , however, is weakly dependent on δT , which though for well characterized systems, i.e., for systems with relative temperature errors that are significantly smaller than $\overline{\delta A_{R,N_2}}/(k_{He} - k_{N_2})$, in practice, is negligible.

Equations (9a) and (10a) also show that the assessments of κ and the $\varepsilon_{0,i}$ s are independent of the penetration depth of the mirrors as well as of gas leakages and outgassing. The reason for the former is that a finite penetration depth affects an assessment in the same way as an error in the temperature does. The reason for the latter is that gas leakages and outgassing affect each data point equally much. The slopes of the fits to the ΔP_i vs P_{Set} plots, k_i s, are thereby not affected; any gas leakage or outgassing by a gas with a refractivity different from that of the gas addressed will, therefore, only contribute with an offset in a ΔP_i vs P_{Set} plot through $\delta P_{l/o,i}$. Note that this comprises leakages and outgassing from the vacuum system and the cavity spacer material, as is the case when He is used in ULE-based cavities,³² as well as virtual leaks from concealed gas volumes in the cavity construction. Leakages and outgassing by gas with the same refractivity will not contribute at all.

Finally, since $(n - 1)_i \propto (1 - \varepsilon_i)$, the systematic errors in $\varepsilon'_{0,i}$'s, i.e., $\delta\varepsilon'_{0,i}$'s, represent the relative errors in the assessment of refractivity of the two gases. Equation (10b) shows that the $\delta\varepsilon'_{0,i}$ are given solely in terms of the errors in the molar polarizabilities of the two gases. Since, when high purity helium is used, $\overline{\delta A_{R,N_2}}$ is larger than $\overline{\delta A_{R,He}}$ by a factor $A'_{R,N_2}/A'_{R,He}$, Eq. (10b) indicates that $\overline{\delta\varepsilon'_{N_2}}$ is smaller than $\overline{\delta\varepsilon'_{He}}$ by the same amount. It also shows that while $\delta\varepsilon'_{0,He}$ is slightly larger than $\overline{\delta A_{R,N_2}}$ (i.e., $1.13 \times \overline{\delta A_{R,N_2}}$), $\delta\varepsilon'_{0,N_2}$ is solely a fraction of it (i.e., $0.13 \times \overline{\delta A_{R,N_2}}$). This shows that the procedure provides such small systematic errors in ε'_{0,N_2} that the cavity deformation solely contributes to the relative error in the assessed refractivity of N_2 by $0.13 \times \overline{\delta A_{R,N_2}}$.

2. Combination of systematic errors of some system parameters

As was alluded to above, the method also provides estimates of a pair of combinations of systematic errors of some system parameters ($\overline{\psi}_i$'s). Regarding these, it is advantageous to separate them into two parts, a (normally dominating) species-independent part, $\overline{\psi}_0$, and a (normally inferior) species-dependent part, $\overline{\delta\psi}_i^s$, i.e., to express $\overline{\psi}_i$ as $\overline{\psi}_0 + \overline{\delta\psi}_i^s$. As is shown by Eq. (SM-41) in the supplementary material,¹⁹ these two can be expressed as

$$\overline{\psi}_0 = \overline{\delta T} - 2\overline{\delta L}_{pd} - \frac{\overline{\delta A}_{R,N_2} - A'_{R,He/N_2} \overline{\delta A}_{R,He}}{1 - A'_{R,He/N_2}} + \overline{\delta P}_{Set} \quad (11a)$$

$$\overline{\delta\psi}_i^s = \frac{A'_{R,He} \overline{\delta A}_{R,N_2} - \overline{\delta A}_{R,He}}{A'_{R,i} (1 - A'_{R,He/N_2})}. \quad (11b)$$

As is shown in Sec. V C of the supplementary material,¹⁹ it is also possible to extract, from the two expressions for k_i s given in Eq. (8), this time by elimination of κ , an expression for an experimentally quantifiable part of $\overline{\psi}_0$, denoted $\overline{\psi}'_0$, that can be expressed as

$$\overline{\psi}'_0 = \frac{k_{N_2} - A'_{R,He/N_2} k_{He}}{1 + k_{N_2} - A'_{R,He/N_2} (1 + k_{He})}. \quad (12)$$

Since this expression provides a value that is virtually indistinguishable from that for $\overline{\psi}_0$, it can be used to assess its value.

As is shown in the supplementary material,¹⁹ the $\overline{\delta\psi}_i^s$'s represent the major parts of the systematic errors in the assessment of $\overline{\psi}_i$'s. They can, therefore, be seen as the systematic errors in the assessed value of the $\overline{\psi}_i$'s.

It is worth to note that the numerator in Eq. (12) represents the deviation from the slopes of the fits that would result if the system would not be affected by any systematic error. Hence, for an ideal system, with no systematic errors, $\overline{\psi}_i$'s would be zero.

D. Assessments of pressure by refractometry and their systematic errors

As is discussed in Sec. V in the supplementary material and as is presented by Eq. (SM-46),¹⁹ when ε'_i 's have been assessed, the refractometer can assess pressure according to

$$P_{Refr,i}^C = \frac{2RT'}{3A'_{R,i}} \left[1 + \xi_{2,i}(T) \frac{\overline{\Delta f} + \overline{\Delta q}}{1 - \overline{\Delta f}} \right] \frac{\overline{\Delta f} + \overline{\Delta q}}{1 - \overline{\Delta f} + \varepsilon'_i} \left(1 + 2\overline{L}'_{pd} \right). \quad (13)$$

Since this pressure is assessed by the use of several measured entities, which, in general, have systematic errors, it will systematically differ slightly from the true one, $P_{True,i}$, by a relative amount, referred to as the relative systematic error of the characterized system, $\overline{\delta P}_{Refr,i}^C$, defined as $(P_{Refr,i}^C - P_{True,i})/P_{True,i}$. As can be deduced from Eq. (SM-46) in the supplementary material,¹⁹ this

can be written as

$$\overline{\delta P}_{Refr,i}^C = \overline{\delta T} - 2\overline{\delta L}_{pd} - \overline{\delta A}_{R,i} - \overline{\delta \varepsilon}'_i + \overline{\delta A}_{R,l/o,i}, \quad (14)$$

where $\overline{\delta A}_{R,l/o,i}$, denoted the gas-leakage-or-outgassing-pressure-referenced relative difference in molar polarizability, is given by $(P_{l/o}/P_{Set})\Delta A_{R,l/o,i}$.

Since the systematic error of the cavity deformation is given by those of the molar polarizabilities [as shown by Eq. (10b)], predominantly by that of nitrogen], $\overline{\delta A}_{R,i}$ and $\overline{\delta \varepsilon}'_i$ are correlated. This implies, as is shown by the Eq. (SM-48) in the supplementary material,¹⁹ that the relative systematic error in the assessment of pressure by use of Eq. (13) can, for both gas species, be expressed as³³

$$\overline{\delta P}_{Refr,i}^C = \overline{\delta T} - 2\overline{\delta L}_{pd} - \frac{\overline{\delta A}_{R,N_2} - \frac{A'_{R,He}}{A'_{R,i}} \overline{\delta A}_{R,He}}{1 - A'_{R,He/N_2}} + \overline{\delta A}_{R,l/o,i}. \quad (15)$$

This shows that when a refractometer is characterized by the cavity deformation assessment procedure presented in this work, the systematic error of a pressure assessment performed by the use of Eq. (13) is affected by the systematic errors in the assessment of the temperature ($\overline{\delta T}$), the penetration depth ($\overline{\delta L}_{pd}$), the molar polarizabilities, primarily that of nitrogen ($\overline{\delta A}_{R,N_2}$) but also, to some extent, that of helium ($\overline{\delta A}_{R,He}$), and the gas-leakage-or-outgassing-pressure-referenced relative difference in molar polarizability ($\overline{\delta A}_{R,l/o,i}$). However, it is not affected by the pressure reference (the pressure reference has in this case solely been used for assessment of the cavity deformation, which could be assessed without influence of the accuracy of the set pressures). This implies that the systematic errors in the pressure reference do not affect the performance of a refractometer characterized by the cavity deformation assessment procedure presented in this work.

It is also worth to note that $\overline{\delta A}_{R,l/o,i}$ is inversely proportional to the set pressure. Hence, its influence on the systematic errors of a pressure assessment decreases the higher the pressure addressed. Moreover, for the case with gas leakages or outgassing into the system by a gas with the same molar polarizability as that addressed (i.e., for which $\overline{\delta A}_{R,l/o,i} = 0$), the systematic error of a pressure assessment is species-independent (it takes the same value for both gas species).

E. Estimates of the uncertainty of the assessed entities

1. Pressure-induced cavity deformation

It is worth noting that since the relative systematic errors in A'_{R,N_2} , T' , and L'_{pd} , i.e., $\overline{\delta A}_{R,N_2}$, $\overline{\delta T}$, and $\overline{\delta L}_{pd}$, are considered to be unknown when assessments are to be performed, they represent relative uncertainties that have systematic origin, here denoted $\overline{u}(A'_{R,N_2})$, $\overline{u}(T')$, and $\overline{u}(L'_{pd})$ respectively. In addition, when the slopes of ΔP_i vs P_{Set} fits, i.e., k_i s, are assessed, they are associated with a given amount of measurement uncertainties, here denoted $u(k_i)$. Since all these entities are uncorrelated, the total uncertainty in the assessment of κ , denoted $u(\kappa')$, can, according to Eq. (SM-49) in

the supplementary material,¹⁹ be written as

$$u(\kappa') = \frac{3A'_{R,He}}{2RT' - 1 - A_{R,He/N_2}} \times \left\{ [\bar{u}(A'_{R,N_2})]^2 + [\bar{u}(A'_{R,He})]^2 + [(k_{He} - k_{N_2})\bar{u}(T')]^2 + [u(k_{He})]^2 + [u(k_{N_2})]^2 \right\}^{1/2}. \quad (16)$$

The corresponding total uncertainties in $\epsilon'_{0,i}$'s, i.e., the $u(\epsilon'_{0,i})$'s, can, according to Eq. (SM-50), be expressed as

$$u(\epsilon'_{0,i}) = \frac{A'_{R,He}}{A'_{R,i} - 1 - A'_{R,He/N_2}} \times \sqrt{[\bar{u}(A'_{R,N_2})]^2 + [\bar{u}(A'_{R,He})]^2 + [u(k_{He})]^2 + [u(k_{N_2})]^2}. \quad (17)$$

2. Combination of systematic errors of some of the system parameters

The total uncertainty in the assessment of the sum of systematic errors, $\bar{\psi}_i$, can similarly, according to Eq. (SM-51), be expressed as

$$u(\bar{\psi}_i) = \frac{A'_{R,He}}{A'_{R,i} - 1 - A_{R,He/N_2}} \times \left\{ [\bar{u}(A'_{R,N_2})]^2 + [\bar{u}(A'_{R,He})]^2 + \left[\frac{A'_{R,i}}{A'_{R,N_2}} u(k_{He}) \right]^2 + \left[\frac{A'_{R,i}}{A'_{R,He}} u(k_{N_2}) \right]^2 \right\}^{1/2}. \quad (18)$$

3. Pressure

When pressure is assessed by a refractometer characterized by the cavity deformation assessment procedure presented in this work, the gas-leakage-or-outgassing-pressure-referenced relative difference in molar polarizability, $\delta A_{R,I/o,i}$, will represent a relative uncertainty by itself, denoted $\bar{u}(A_{R,I/o,i})$. This implies that when pressure is assessed by the use of Eq. (13), the relative uncertainty from the cavity deformation assessment process can, as is shown by Eq. (SM-54) in the supplementary material,¹⁹ be expressed as

$$\bar{u}(P_{Ref,i}^C) = \left\{ \left[\frac{\bar{u}(A'_{R,N_2})}{1 - A'_{R,He/N_2}} \right]^2 + \left[\frac{\bar{u}(A'_{R,He})}{1 - A'_{R,He/N_2}} \right]^2 + [\bar{u}(T')]^2 + [\bar{u}(\bar{I}_{pd})]^2 + [\bar{u}(A_{R,I/o,i})]^2 + \left(\frac{A'_{R,He}}{A'_{R,i} - 1 - A'_{R,He/N_2}} \right)^2 \{ [u(k_{He})]^2 + [u(k_{N_2})]^2 \} \right\}^{1/2}. \quad (19)$$

Since assessments of pressure can be performed with an absolute (total) accuracy, $u(P_{Ref,i}^C)$, that is given by

$$u(P_{Ref,i}^C) = \sqrt{[u(P_{min})]^2 + [\bar{u}(P_{Ref,i}^C)P]^2}, \quad (20)$$

where $u(P_{min})$ denotes the minimum detectable pressure of the refractometer system under scrutiny and $\bar{u}(P_{Ref,i}^C)$ represents the relative uncertainty from the deformation assessment process, given by Eq. (20), this implies that the total accuracy of the assessment of pressure, $u(P_{Ref,i}^C)$, is given by $\bar{u}(P_{Ref,i}^C)P$ for the case when the pressure is larger than $u(P_{min})/\bar{u}(P_{Ref,i}^C)$. Since $u(P_{min})$ can take values down to the tens of μPa range,¹⁸ for the case when $\bar{u}(P_{Ref,i}^C)$ is in the tens or hundreds of ppm range, this takes place when pressures above a few Pa range are assessed. Under these conditions, the relative total accuracy of the pressure assessment is given by Eq. (19).

IV. EXPERIMENT

A. Instrumentation

To demonstrate the execution and applicability of this novel procedure for assessment of cavity deformation in FP-based refractometry systems, it was applied to a previously developed Invar-based FP cavity described elsewhere.¹⁸ In short, it comprises a DFPC machined from a 70 mm round stock of Invar resulting in a $150 \times 70 \times 50$ mm block in which two cavities with diameters of 6 mm were drilled.³⁴ Each cavity is made of two 12.7 mm concave mirrors with a radius of curvature of 500 mm and a reflectivity of 99.97% (for detailed information about the instrumentation and the various parts used, see Ref. 18), yielding cavities with a finesse of 10^4 and FSRs of around 1 GHz. As is shown in Fig. 2, the mirrors are not permanently fused to the spacer; instead they are clamped directly onto the cavity spacer by the use of back plates and O-rings. This implies that they can easily be removed, cleaned, or replaced.

The DFPC is placed inside a temperature stabilized aluminum enclosure (oven). The temperature of the cavity spacer is monitored by three Pt-100 sensors, which, at room temperature, have a standard calibration uncertainty of 200 mK,³⁵ mounted in holes drilled in the spacer between the two cavities.

Each cavity is probed by laser light produced by an Er-doped fiber laser emitting light at around 1550.14 nm that is coupled into an acousto-optic modulator (AOM) for fast frequency compensation for the frequency locking process. The frequency shifted first order output of the AOM is, in turn, coupled into a 90/10 fiber splitter. Although the laser is continuously tunable by a built-in piezo crystal about 6 GHz (~ 0.05 nm), it was typically scanned only slightly more than 1 FSR (~ 0.01 nm) before it made a controlled mode jump (see below).

To lock the laser to a cavity mode, the light from the 90% output of the fiber splitter is sent to an electro-optic modulator

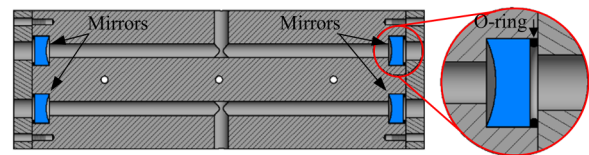


FIG. 2. Schematic illustration of the DFPC assembly under scrutiny. The mirrors are pressed against the Invar spacer by O-rings compressed by back plates. The white circles are holes for temperature sensors.

(EOM) that is modulated at 12.5 MHz for Pound–Drever–Hall (PDH) locking. The output of the EOM is sent through an optical circulator to a collimator whose output is mode-matched to the TEM₀₀ mode of the FP cavity. The back reflected light is picked up by the collimator and routed through the circulator onto a fast photo detector, while the light transmitted through the cavity is monitored by a large area photo detector. For each arm, the outputs from the photo detectors are connected to a field programmable gate array (FPGA). In this, the signal from the reflection detector is demodulated at 12.5 MHz to produce the PDH error signal. The slow components of the feedback (<100 Hz) are sent to the laser, while the fast ones (>100 Hz) are sent to a voltage controlled oscillator that produces an RF signal that drives the AOM at 110 MHz.

To sample the beat frequency, the light from the 10% fiber splitter outputs of the two arms are combined in a 50/50 fiber coupler. This light is sent to a fiber-coupled photo detector whose RF signal is measured by a frequency counter. The frequency of the laser light is assessed (with an uncertainty of 2×10^{-7}) by the use of a wavemeter.

To accommodate large shifts in cavity mode frequencies, the system comprises an automatic relocking routine. When the feedback to a laser falls below or exceeds some preset levels, the laser makes a controlled mode jump. In this process, the signal from the transmission detector is used to modulate the strength of the feedback sent to the laser and the AOM. To determine the number of mode jumps made, Δq , the feedback voltages sent to the lasers are monitored with an analog input module.

To provide the system with a reference pressure a dead weight pressure balance was connected to the system. Its set pressure, P_{Set} , was calculated as³⁶

$$P_{\text{Set}} = \frac{(m + m_p)g}{A_p[1 + \alpha(T_p - T_{p,0})]} + P_R, \quad (21)$$

where m is the mass added to the balance, m_p is the mass of the piston, g is the local gravity, A_p is the effective piston area, α the area thermal expansion coefficient, T_p is the piston temperature, $T_{p,0}$ is the temperature at which A_p was measured, and P_R is the hood reference pressure. The hood was continuously pumped down with a molecular turbo pump and measured with a pressure gauge. The piston temperature, $T_{p,0}$, was continuously monitored with a standard Pt-100 sensor.

B. Methodology

To reduce the influence of fluctuation and drifts, the assessments were performed by the GAMOR methodology. It has previously been shown that this methodology has the ability to reduce the influence of fluctuations and drifts on refractometry assessments considerably, including the effect of leakages and outgassing.^{12,13} It has recently allowed for realizing high-precision (sub-ppm or sub-mPa) DFPC-based refractometry in metallic cavities.¹⁸ As was alluded to above, its extraordinary abilities have been attributed to a measurement procedure based on two cornerstones, viz.,

- (i) the refractivity of the gas in the measurement cavity is assessed by the use of a rapid gas modulation procedure that allows for a repetitive referencing of the filled measurement cavity beat frequency assessments to empty cavity assessments¹⁵ and
- (ii) the empty measurement cavity beat frequency at the time when the filled measurement is probed is estimated based on an interpolation between two empty measurement cavity beat frequency measurements taken just prior to and directly after the filled measurement cavity measurements.

The construction of the cavity system, the modulation procedures, and the running conditions have all been chosen so that the system is not expected to be noticeably affected by any Joule heating or any finite stabilization time of the pressure reference and to provide stable temperature conditions.³⁷

C. Experimental procedure

The assessment of cavity deformation in the system described above was made for five set pressures, ranging from 2 841 to 15 997 Pa, for two gases, N₂ and He. In order to reduce the risk of gas contamination, the measurements were made in two separate series of which those made on N₂ were taken before those on He. Moreover, in order to minimize the effect of drifts, the assessments of the five pressures were made in a nonconsecutive order, viz., 4 302.8, 8 687.7, 15 996.7, 2 841.2, and 11 610.6 Pa.

For each set pressure and for each gas, 100 consecutive gas fillings and evacuation cycles were made, producing in total 1000 data points. Each gas filling-and-evacuation cycle took 100 s.

V. RESULTS

Figure 3 presents a set of data points in the form of individual markers. The upper panel shows two sets of measurement data taken for five dissimilar set pressures of He (upper data set, red markers) and N₂ (lower set, blue markers) in terms of the difference between the pressure assessed using the uncharacterized refractometer, $P_{\text{Refr},i}^U$, and the pressure set by the pressure reference system, P_{Set} , ΔP_i , as a function of the set pressure.

The straight lines represent fits of linear functions of the type $a_i + k_i \times P_{\text{Set}}$, where a_i s represent the gas leakage and outgassing into the system together with the offset in set pressure, while k_i 's represent k_{He} and k_{N_2} , defined above.

For the case when the evaluation is based on the molar polarizability for N₂ that is traceable to a thermodynamic standard, k_{He} and k_{N_2} were assessed to 0.016 317(3) and 0.001 611(2), respectively, where the numbers in parentheses (3×10^{-6} and 2×10^{-6}) represent their ($k = 2$) measurement uncertainties, $u(k_i)$. The two offset values, a_i s, were found to be, for He and N₂, 1.44(3) and −0.03(2) Pa, respectively. The lower panel shows the residual between the data taken and the fits.

For the case when the evaluation is based on the molar polarizability for N₂ that is traceable to a mechanical standard, k_{N_2} changes to 0.001 610(2) while k_{He} and the offset values remain the same.³⁸

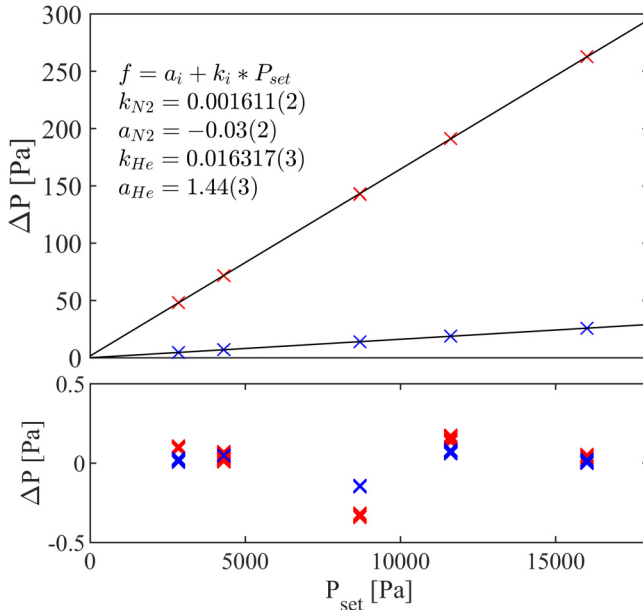


FIG. 3. Top panel: \times —markers: The difference in pressure of the uncharacterized refractometer and the set reference pressure of the pressure balance, ΔP_i , as a function of set pressure for He (upper data set, red markers) and N₂ (lower data set, blue markers). Straight lines: linear fits to the data points. Bottom panel: The residual pressure with respect to the linear fits. Note that each of the ten groups of markers in the two panels comprises 100 independent measurements whose spread in panel a is smaller than the size of the markers. The evaluation of the data is based on the molar polarizability for N₂ given in Table I that is traceable to a thermodynamic standard. For the case when the evaluation is based on the molar polarizability traceable to a mechanical standard, k_{N_2} changes to 0.001 610(2) while k_{He} remains the same.

A. Assessment of the pressure-induced cavity deformation

When the evaluation is based on the molar polarizability that is traceable to a thermodynamic pressure standard, i.e., using the values for the slopes of the fits that are given in Fig. 3, together with the expressions for the deformation parameters from above, i.e., Eqs. (9a) and (10a), gives, with T' taken as 296.15 K and $A'_{R,He/N_2}$ as 0.117 761 8(37), values for κ' of $5.2575 \times 10^{-12} \text{ Pa}^{-1}$ and the two ϵ'_i 's of 16.669×10^{-3} and 1.9630×10^{-3} for He and N₂, respectively.

The uncertainties of these values are given by Eqs. (16) and (17). When evaluating these, it can first be noted that, since the helium used solely had a purity of 0.999 999, it can contribute with a relative uncertainty in the molar polarizability of helium of 10 ppm. According to measurement data, $u(k_{He})$ and $u(k_{N_2})$ take values of 3×10^{-6} and 2×10^{-6} , respectively.

Moreover, in this case, the uncertainty in the molar polarizability of N₂, $\bar{u}(A'_{R,N_2})$, takes, according to Table I, a value of 31 ppm. In addition, when the uncertainty in κ is to be assessed, the $[(k_{He} - k_{N_2})\bar{u}(T')]$ term needs to be estimated. Using the standard ($k = 2$) uncertainty of the temperature sensors (200 mK)³⁵ as

an estimate of $\bar{u}(T')$ implies that this term takes a value of 10 ppm. This implies that the uncertainty in κ' , ϵ_{He} , and ϵ_{N_2} , i.e., $u(\kappa')$, $u(\epsilon_{He})$, and $u(\epsilon_{N_2})$, become $12 \times 10^{-15} \text{ Pa}^{-1}$, 38×10^{-6} , and 4×10^{-6} , respectively. These values represent relative uncertainties of 2.3% for κ and ϵ'_{He} and 2% for ϵ'_{N_2} 's.

For the case when the evaluation is based on the molar polarizability that is traceable to a mechanical pressure standard, for which k_{N_2} takes a value of 0.001 610(2), i.e., Eqs. (9a) and (10a), gives, with $A'_{R,He/N_2}$ taking a value of 0.117 761 7(9), values for κ' of $5.2577 \times 10^{-12} \text{ Pa}^{-1}$ and the two ϵ'_i 's of 16.670×10^{-3} and 1.9631×10^{-3} for He and N₂, respectively.

In this case, the uncertainty in the molar polarizability of N₂, $\bar{u}(A'_{R,N_2})$, takes a value of 8 ppm. The corresponding numbers for $u(\kappa')$, $u(\epsilon_{He})$, and $u(\epsilon_{N_2})$ then become $5.9 \times 10^{-15} \text{ Pa}^{-1}$, 15×10^{-6} , and 2×10^{-6} , respectively, while the relative uncertainties become, in all cases, 1%.

Since refractivity is proportional to $1 - \epsilon_i$, an uncertainty of ϵ_{N_2} in the 10^{-6} range implies, in turn, that a system utilizing the presented procedure, even if the cavity deformation assessment was performed with a helium quality giving rise to a relative uncertainty in the molar polarizability of helium of 10 ppm, should be able to assess refractivity of N₂ with a relative accuracy in the low ppm range; solely 4 and 2 ppm when the deformation assessment is traceable to a thermodynamic and mechanical standard, respectively.

B. Assessment of the combinations of species-independent systematic errors

Using the values for the slopes of the fits, i.e., the k_i 's, the species-independent part of $\bar{\Psi}_i$, i.e., $\bar{\Psi}'_0$, could, by the use of Eq. (12), similarly be assessed to -353×10^{-6} . The magnitude of this value is expected to not exceed the combined uncertainty of all entities in Eq. (7). In our case, it is well within the estimated combined uncertainties of the system; the uncertainty of the type A Pt-100 sensors used is (200 mK) 700 ppm,³⁵ the uncertainty of the set pressure is 40 ppm, and the uncertainties of the molar polarizabilities for He, which, for the helium gas used in the work, is 10 ppm, and N₂ which is 8 or 31 ppm. To improve on this situation, work with lowering the uncertainty of the temperature assessment and the use of helium gas with higher purity is presently under way and will be reported elsewhere.

C. Offsets—Indications of gas leakage or outgassing into the system that the cavity deformation assessment procedure is insensitive to

The a_i parameters in the fits represent gas leaking and outgassing into the cavities and the absolute offset error in the set pressure. The value of $-0.03(2) \text{ Pa}$ for a_{N_2} indicates that the N₂ assessment is not significantly affected by such processes. The value of a_{He} , however, is markedly higher [1.44(3) Pa]. This difference indicates that there is a small leakage into the measurement cavity.

However, since the deformation assessment procedure developed here makes use of the slopes of the fits, which are assumed to be unaffected by any offset in a ΔP_i vs P_{Set} plot (i.e., any nonzero value of the a_i parameter), it is considered that this gas leakage or outgassing will not affect the cavity deformation assessment noticeably.

VI. SUMMARY, DISCUSSION, AND CONCLUSIONS

A. Procedure

In this work, we present a novel procedure for robust assessment of cavity deformation in FP refractometers that provides small amounts of uncertainty. It is inspired by a method proposed by Egan *et al.* in which one gas, whose molar polarizability is known (e.g., He), is used to characterize the system with regard to its pressure-induced deformation, allowing for subsequent accurate assessments of the pressure of other gases.¹⁴ The procedure presented differs though from that suggested by Egan *et al.* by the fact that it assesses the difference between the pressure assessed by the uncharacterized refractometer and a supplied reference pressure at a series of pressures, i.e., as ΔP_i vs P_{Set} , for two gases with dissimilar refractivity (here He and N₂). By then fitting linear curves to these responses, their slopes are used as a basis for the assessment of the pressure-induced deformation of the cavity with no (or a minimum of) influences from the systematic errors or uncertainties in the assessment of gas temperature, estimates of penetration depth, offsets, and linear errors in the pressure reference used, and, if the system is utilizing a fixed length of the gas filling cycle, as tacitly is assumed in the GAMOR methodology, gas leakages or outgassing into the system.

To demonstrate this, explicit expressions for the assessment of two pressure-induced cavity deformation parameters, κ [defined as $(\delta L/L_0)/P$] and ε_i [defined as $(\delta L/L_0)/(n-1)_i$], have been derived. It was found that, when a pressure reference with a species-independent response is used (i.e. one for which $\delta P_{Set,He} = \delta P_{Set,N_2}$), both deformation parameters can be determined independently of the systematic error in the set pressure, while ε_i can be determined also independently of the systematic error in the assessment of temperature. Since outgassing and gas leaking into the cavities predominantly contribute with an offset in the ΔP_i vs P_{Set} responses, the procedure will, as long as the GAMOR methodology is used, also not be significantly affected by such processes.

The procedure can be carried out for any “secondary” gas, irrespective of the uncertainty in its molar polarizability. The analysis indicates that in the proposed procedure, if carried out on a high-precision refractometry system, ε_i entity for He, ε_{He} , “inherits” the relative uncertainty in the molar polarizability of the “other gas,” while the same entity for this gas when N₂ is addressed, i.e., ε_{N_2} is, when high purity helium is used [so that $\bar{u}(A'_{R,He})$ can be neglected with respect to $\bar{u}(A'_{R,N_2})$], affected significantly less by this (solely by 13% of it).

B. Assessment of cavity deformation of a specific FP-based cavity

To illustrate the execution and applicability of the procedure presented, we have assessed the cavity deformation of a previously developed refractometry system based on a dual FP cavity made of Invar operating at 296.15 K and 1550.14 nm.¹⁸ Under the condition that the set pressure from the reference system is species-independent, but taking into account that the purity of the helium used provided an uncertainty in $A_{R,He}$ of 10 ppm, κ could be assessed with a relative uncertainty of 2.3‰ or 1‰ to $5.258(12) \times 10^{-12}$ and $5.258(6) \times 10^{-12}$ Pa⁻¹ (when the molar polarizability of N₂ is traceable to a

thermodynamic and a mechanical standard, respectively). The two ε_i 's could, for He, be assessed with similar relative uncertainties, 2.3‰ or 1‰, to $16.669(38) \times 10^{-3}$ and $16.670(15) \times 10^{-3}$ and likewise, for N₂, with 2‰ or 1‰ uncertainties to $1.9630(4) \times 10^{-3}$ and $1.9631(2) \times 10^{-3}$ (for the same two cases). The key reason the relative uncertainties of the deformation parameters could be assessed at low ‰-levels, for ε_{N_2} corresponding to low 10⁻⁶-levels on an absolute scale, is that the high precision of the system scrutinized¹⁸ allows for linear regressions of the ΔP_i vs P_{Set} plots with uncertainties [$u(k_{N_2})$ and $u(k_{He})$] that are smaller than the present relative uncertainty of the molar polarizability of N₂.

This shows that irrespective of which value of the molar polarizability for N₂ that is used for the cavity deformation assessment, the deformation parameters take values that overlap well within their uncertainties. The dissimilar uncertainties of the molar polarizabilities only provide various degrees of uncertainty of the deformations. It is particularly noticeable that the procedure provides an uncertainty in the deformation parameter ε_i that, when nitrogen is assessed, is in the low 10⁻⁶-level (4×10^{-6} and 2×10^{-6} , respectively). Since refractivity is proportional to $1 - \varepsilon_i$, an uncertainty of ε_{N_2} in the 10⁻⁶ range implies that a system utilizing the presented procedure should be able to assess refractivity of N₂ with a relative accuracy in the low ppm range.

C. Conclusions and outlook

The work has thus shown that, for the case when the refractometer system to be characterized has a high precision (so that it can provide measurement uncertainties of the slopes of the fits in ΔP_i vs P_{Set} plots in the low 10⁻⁶ range) and when high purity helium is used [so $\bar{u}(A'_{R,He})$ can be neglected with respect to $\bar{u}(A'_{R,N_2})$], the procedure presented is capable of assessing cavity deformation, ε'_{N_2} , to within a fraction of the uncertainty in the molar polarizability of nitrogen. For the pertinent experimental situation, i.e., a wavelength of 1550.14 nm and a temperature of 296.15 K, the deformation can be assessed with such high accuracy that its uncertainty contributes to the assessment of pressure of N₂ solely by 4 and 1 ppm, for the cases when polarizabilities are traceable to a thermodynamic and a mechanical standard, respectively.

The fact that the uncertainties of the slopes of the fits are smaller than those originating from systematic errors implies that the assessments of the deformation parameters and the refractivity cannot be significantly improved by an improved measurement processes; their accuracies would benefit mostly from an improved accuracy of the molar polarizability of N₂ under the prevalent conditions (at the pertinent wavelength and temperature).

Since pressure, when assessed by a refractometer, is affected by the uncertainties in a number of entities, among them the molar polarizability of the gas addressed, this implies that when a system whose cavity deformation has been assessed by the presented procedure is to be used for assessment of pressure or as a pressure standard, cavity deformation will play an inferior role (it is no longer a limiting factor). As long as the temperature can be assessed with an uncertainty that is in the low ppm region, if the system has a negligible amount of leakage and outgassing, and if high purity gases are used, it would allow for assessment of pressure or

realization of the Pascal with an uncertainty that is limited by the accuracy of the molar polarizability of the gas addressed.

It is important to emphasize that, although the procedure presented for determination of the deformation was made in a limited pressure range (up to 16 kPa), Eq. (13) is valid for the entire range of pressures for which Eqs. (2) and (3) are valid. If higher pressures need to be assessed, expressions similar to Eq. (13), based upon expanded expressions for the density [Eq. (2)] and pressure [Eq. (3)], that also include higher order virial coefficients,^{3,14,39} should be utilized.

It can finally be concluded that if the molar polarizability of a second gas, e.g., Ne, is determined by *ab initio* calculations with higher accuracy than by which the molar polarizability of nitrogen presently is known, Eqs. (9b) and (10b) indicate that the deformation (κ' as well as the ε'_i s) could be assessed with even higher accuracy. In this case, Eq. (19) indicates that, for a system with a low uncertainty in the temperature assessment, the accuracy of the assessment of pressure could be improved beyond that predicted by the He/N₂ system analyzed in this work.

ACKNOWLEDGMENTS

This research was, in part, supported by the EMPIR Initiative (No. 18SIB04), which is cofounded by the European Union's Horizon 2020 research and innovation program and the EMPIR Participating States; the Swedish Research Council (VR) (Project No. 621-2015-04374); the Umeå University Industrial Doctoral School (IDS); the Vinnova Metrology Programme (Project Nos. 2018-04570 and 2019-05029); and the Kempe Foundations (Project No. 1823, U12).

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- ¹⁶Note that we by the phrase gas leakages and outgassing into the system (or sometimes solely gas leakages and outgassing) mean leakages or outgassing of gas into the system that, during each gas modulation cycle, continuously and monotonically, change the composition of the gas. This phrase does thereby not

include any contamination of the gas supply, which provides a static level of impurities.

¹⁷We will here, by the expression “systematic errors,” denote errors that are constant over an appreciable time (at least over the time over which the deformation assessment takes place and the assessment should be considered to be valid).

¹⁸I. Silander, C. Forssén, J. Zakrisson, M. Zelan, and O. Axner, *Opt. Lett.* **45**, 2652 (2020).

¹⁹See supplementary material at <https://doi.org/10.1116/6.0000375> for derivations of expressions that relate the change in beat frequency to the refractivity of the gas in DFPC-based refractometry, an assessment of the conditions for when the virial coefficient terms can be neglected when deformation and refractivity are assessed, a compilation of molecular data for the gases under scrutiny, a derivation of expressions for gas density and pressure in terms of refractivity, a detailed description of the novel procedure for the assessment of cavity deformation of refractometer systems and assessment of pressure, some comments on the need to assess the length of the cavity, and nomenclature.

²⁰The deformation comprises both changes in the length of the cavity spacer and deformation of the mirrors. For the case when a DFPC is used, it represents the difference in length deformation of the two cavities when gas is filled into one of them.

²¹O. Axner, I. Silander, T. Hausmaninger, and M. Zelan, “Drift-free Fabry-Pérot-cavity-based optical refractometry—Accurate expressions for assessments of gas refractivity and density,” e-print [arXiv: 1704.01187v2](https://arxiv.org/abs/1704.01187v2) (2017).

²²The omitted third order density virial term contributes to the density of nitrogen with 1 ppm for a refractivity of 1.5×10^{-3} , which corresponds to, under room temperature conditions, a pressure of 5.6×10^5 Pa, far above that used in the present cavity deformation assessment.

²³M. Puchalski, K. Piszczatowski, J. Komasa, B. Jeziorski, and K. Szalewicz, *Phys. Rev. A* **93**, 032515 (2016).

²⁴The omitted third order pressure virial term contributes to the pressure of nitrogen with 1 ppm for a density of N₂ of 870 mol/m³, representing, under room temperature conditions, a pressure of 20×10^5 Pa, far above that used in the present deformation assessment.

²⁵A. Rizzo, C. Hättig, B. Fernández, and H. Koch, *J. Chem. Phys.* **117**, 2609 (2002).

²⁶NIST, “The NIST reference on constants, units, and uncertainty,” <https://physics.nist.gov/cgi-bin/cuu/Value?r> (accessed August 20, 2020).

²⁷An alternative means to assess q_0 is as the closest integer to the ratio of the frequency of the laser addressing the evacuated measurement cavity, ν_0 , and the FSR of the cavity.

²⁸Since the deformation assessment is done at pressures for which the influence of the nonlinear terms of Eqs. (2)–(4) plays an insignificant role, no systematic errors of the virial coefficients need to be considered.

²⁹J. W. Schmidt, S. A. Tison, and C. D. Ehrlich, *Metrologia* **36**, 565 (1999).

³⁰While $\delta P_{Set,i}$ is assumed to be proportional to the pressure, δP_{Set}^0 , $\delta A_{R,i}$, δT , and δL_{pd} are assumed to be pressure-independent constants.

³¹Since, as is shown in Sec. III of the supplementary material, the nonlinear part of ε_i (i.e., $\varepsilon_{B,i}$) also can be expressed (by the use of virial coefficients) in terms of $\varepsilon_{0,i}$, an assessment of $\varepsilon_{0,i}$ provides an estimate of ε_i for the entire range of pressures over which Eq. (4) is valid.

³²Y. Yang and T. Rubin, *J. Phys. Conf. Ser.* **1065**, 162003 (2018).

³³It should be noted that in Eq. (15) there are no explicit (or additional) contributions from the cavity deformation to the systematic errors of the pressure assessment.

³⁴The advantages of this cavity type compared with classical cavities made from low thermal expansion glass, e.g., Zerodur or ULE, are (1) a higher thermal conductivity, which allows for more accurate temperature assessments; (2) a higher Young's modulus, which gives the cavity a lower pressure-induced deformation; (3) a lower gas diffusivity, permeation, and solubility of He; and (4) lower fabrication costs.

³⁵IEC 60751:2008, *Industrial platinum resistance thermometers and platinum temperature sensors*, International Electrotechnical Commission, 2nd ed. (2008).

³⁶C. M. Sutton, *Metrologia* **30**, 591 (1994).

³⁷GAMOR performed in a narrow-channel cavity, as is the case for the Invar-based refractometer,¹⁸ and at the relatively low pressures used in this work (up to 6 kPa), is not significantly affected by any pV-work. Since the Joule-Thomson coefficient, $\mu_J T$, which is defined as $(\delta T / \delta P)_H$, has, for N₂, a value of 200 mK/bar, the filling of gas in the assessments performed in this work (up to 16 kPa) is expected to give a change in gas temperature of up to 32 mK. In addition, for the setup used, there is a 10 cm long small diameter (4 mm) metallic tube between the valve and the cavity. The gas will, therefore, by collisions with the tube wall, to a large degree equilibrate in temperature with respect to that of the tube (which has the same temperature as the cavity spacer) before it enters the cavity. Moreover, since the semidiameter of the cavity is small (3 mm) and the pressure is restricted (below 16 kPa), the diffusion time of gas from the center of the cavity to the cavity walls is in the sub-second range. This implies that there will be a rapid equilibration also between any residual temperature difference between the gas and the cavity walls. Finally, since the gas has a significantly smaller heat capacity than the cavity spacer material and the cavity volume is small (4 cm³), there will (for any remaining minute residual difference in temperature between the gas when it

enters the cavity and the cavity wall temperature), not be any significant change of the temperature of the cavity wall because of the gas-wall collisions. We can, therefore, also assume that the cavity wall temperature will not be affected significantly by the gas filled into the cavity. Since the thermal conductivity in the cavity spacer is reasonably high, we do not expect any systematic deviations between the temperatures of the probes and that of the cavity walls. This implies that the temperature of the gas in the laser beam at the time of the assessment of refractivity is not expected to deviate from that assessed by the temperature probes by more than the uncertainty of the calibration of the probes. The system is also not noticeably affected by the finite settling time of the dead weight pressure balance. The length of the gas filling-and-emptying cycle was chosen so as to, for the pressure range addressed, not be noticeably affected by any of these processes, but also as short as possible to minimize the influence of white noise during a given campaign time.

³⁸For the pressure range utilized in this work, i.e., for pressures in the 2841 to 15 997 Pa range, the pressure-induced cavity deformation shows no nonlinearity. This supports the approximation made in the last step of Eq. (6).

³⁹H. J. Achterman, T. K. Bose, M. Jaeschke, and J. M. St-Arnaud, *Int. J. Thermophys.* **7**, 357 (1986).