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Invar-based refractometer for pressure assessments

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Gas modulation refractometry (GAMOR) is a methodology that can mitigate fluctuations and drifts in refractometry. This can open up for the use of non-conventional cavity spacer materials. In this paper, we report a dual-cavity system based on Invar that shows better precision for assessment of pressure than a similar system based on Zerodur. This refractometer shows for empty cavity measurements, up to 10^4 s, a white noise response (for N_2) of $3 \text{ mPa s}^{1/2}$. At 4303 Pa, the system has a minimum Allan deviation of 0.34 mPa (0.08 ppm) and a long-term stability (24 h) of 0.7 mPa. This shows that the GAMOR methodology allows for the use of alternative cavity materials. © 2020 Optical Society of America

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In the SI system of units, the Pascal is defined as force per unit area. In practice, it is realized with mechanical devices such as pressure balances and liquid manometers [1–5]. With the revision of the SI system, an alternative path to realize the Pascal became feasible [6]. By measuring the refractivity and the temperature of a gas, it is possible to calculate its pressure by the use of the Lorentz–Lorenz equation and an equation of state [7–11]. Such a realization of the Pascal does not depend on any mechanical actuator but instead measures directly the gas density, thereby potentially decreasing uncertainties and shortening calibration chains.

The most sensitive refractometers are based on Fabry–Perot (FP) cavities where a laser is used to probe the frequency of a longitudinal mode [12–19]. Although such refractometers have the potential to provide highly accurate measurements, their cavities, usually bored in low thermal expansion glass, e.g., Zerodur or ultra-low expansion glass (ULE), are sensitive to mechanical disturbances and long-term length drifts [10,20,21].

To alleviate some of these limitations the authors have recently developed a new type of methodology, called gas modulation refractometry (GAMOR), in which the measurement cavity is repeatedly filled and emptied with gas [18,19]. While classical FP-based refractometry requires cavity length stability over long measurement periods, typically days, GAMOR refractometers rely on a length stability solely over individual

gas modulation cycles, typically around 100 s. This significantly reduces the pick-up of fluctuations and drifts of the cavity length on time scales longer than the measurement cycle. It also reduces the effects of gas leaks, gas permeation, and outgassing [18,19].

These less stringent requirements for GAMOR refractometry open up for the use of alternative cavity spacer materials. One such is Invar, which, compared to commonly used glass materials, has a number of advantages, mainly: (1) a five times higher ratio of thermal conductivity to volumetric heat capacity, which implies that the cavity will have lower thermal gradients, which, in turn, allows for more accurate temperature assessments; (2) a two times higher volumetric heat capacity, which reduces temperature fluctuations of the cavity, (3) a 50% higher Young's modulus, which gives the cavity a lower pressure induced deformation; (4) potentially a lower degree of He diffusivity, permeation, and solubility compared to some of the glass materials [22]; and (5) it can be machined in a standard metal workshop, whereby more complicated geometries can be created more swiftly at a lower cost.

In this Letter, we describe a GAMOR system based on an Invar cavity. We start by describing the experimental setup. To evaluate the suitability of Invar as an alternative cavity spacer material, we then characterize the system and compare it to previously published results from a Zerodur cavity.

The refractometer is based on a dual-FP-cavity (DFPC). The refractivity of the gas under scrutiny, let into one of the cavities, the measurement cavity, can be expressed as a function of the shift of the beat frequency between the two cavities, Δf , the shifts in mode numbers of the modes addressed in the two cavities, Δq_1 and Δq_2 , and a normalized relative cavity deformation, ε , as

$$n - 1 = \frac{\overline{\Delta f} + \overline{\Delta q_1}}{1 - \overline{\Delta f} + \varepsilon}, \quad (1)$$

where $\overline{\Delta f}$ is the shift of the relative beat frequency, adjusted for possible mode jumps in the reference cavity, given by $(\Delta f - \nu_{02}\Delta q_2/q_{02})/\nu_{01}$, where ν_{01} and ν_{02} are the empty cavity frequencies of the two lasers, while q_{02} is the number of the mode addressed in the reference cavity. $\overline{\Delta q_1}$ is a shorthand notation for $\Delta q_1/q_{01}$, where q_{01} is the number of the mode addressed in the empty measurement cavity. ε is given by

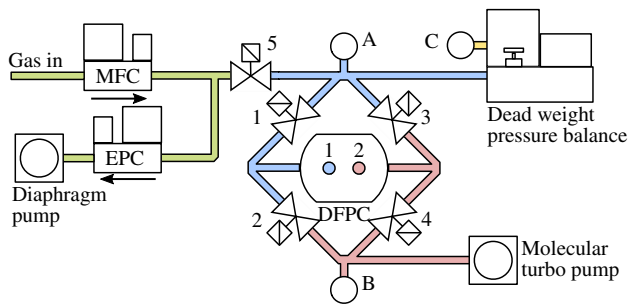


Fig. 2. Schematic illustration of the vacuum system. The gas supply part is shown in green, the part where the pressure is assessed is displayed in blue, and the evacuation part is illustrated in red.

state, resulting in a regulated pressure. By simultaneously monitoring the hood pressure with pressure gauge C and the piston temperature (averaged over 5 min.), the actual pressure set by the pressure balance, P_{Set} , can then be estimated.

The gas modulation is achieved by switching the valves between two states. State I, gas filling, is initiated by opening valves 1 and 4, whereby cavity 1 is filled with gas, while cavity 2 is evacuated. The length of this state is chosen so that the pressure in the measurement cavity stabilizes to that provided by the pressure balance (50 s). In state II, valves 2 and 4 are open, while valve 1 is closed, whereby both cavities are being evacuated (50 s). The valves are controlled through a digital output module (National Instruments, NI-9474).

All data are evaluated by the use of the interpolation methodology that is a part of the GAMOR methodology. In this methodology, the shift in the beat frequency, Δf , is taken as the difference between the mean value of the beat frequency, f , measured over the last 10 s of state I (i.e., with gas in the measurement cavity) and the interpolated value of two empty cavity beat frequency measurements (state II), each averaged over 5 s [18,19].

The measurement campaign starts by assessment of the frequencies of the two lasers when locked to evacuated cavities, i.e., ν_{01} and ν_{02} , which, by the use of a wavemeter (Burleigh, WA-1500) are assessed with an uncertainty of 2×10^{-7} , and the mode numbers addressed for empty cavities, i.e., q_{01} and q_{02} , are assessed uniquely (i.e., with no uncertainty).

Furthermore, the DFPC has recently been preliminary characterized [25]. It was found that the normalized relative cavity deformation for the measurement cavity for N_2 , ε , is $1.968(1) \times 10^{-3}$. The characterization also provided a deformation independent correction term, ψ , that originates from a combination of systematic errors in the assessment of the temperature, the molecular polarizability of N_2 , and the pressure from the pressure balance, which amounts to $-373.1(1) \times 10^{-6}$. The main contribution to this is attributed to the temperature assessment [it is in parity with the standard uncertainty of the Pt-100 sensors used (200 mK)].

To evaluate the performance of the system, a pair of long-term measurements on N_2 at 296.15 K were performed, one with the pressure balance set to 4303 Pa and one in which no gas was supplied to the refractometer. Figure 3 shows a set of measurement data taken over 24 h [26], presented as the difference, ΔP , between the pressure measured by the refractometer, P_{Ref} , assessed from Eqs. (1)–(3) using molecular parameter values

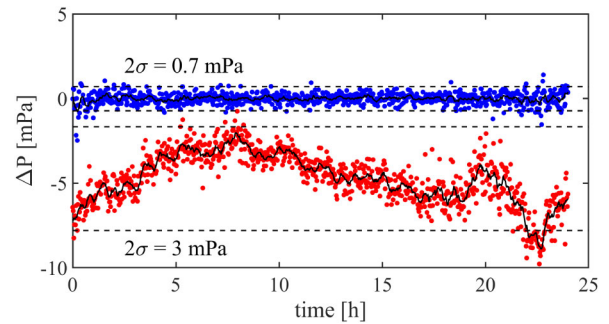


Fig. 3. Difference between the pressure of N_2 measured by the refractometer (corrected by ψ), P_{Ref} , and the estimated pressure supplied to the refractometer, P_{Set} , denoted ΔP , for an empty cavity (blue) and at 4303 Pa (red), respectively. The black curves represent moving averages of 10 samples. The dashed lines correspond to $\pm 2\sigma$ of the assessed pressure difference.

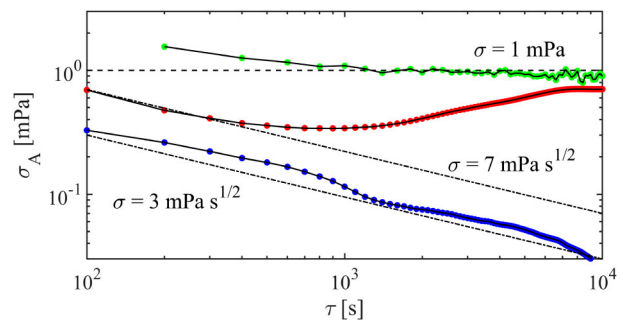


Fig. 4. Allan deviations, σ_A , of pressure assessments made by the GAMOR methodology. Green markers: data previously obtained at 4303 Pa from a Zerodur cavity [19]; red markers: data taken at the same pressure by the Invar-based system presented in this work; blue markers: data taken by the Invar-based system with an empty measurement cavity; dashed horizontal line: Allan deviation of 1 mPa; dashed-dotted slanting lines: Allan deviations corresponding to a white noise level of 7 and 3 $\text{mPa s}^{1/2}$, respectively.

from [18], corrected by ψ , and the estimated set-pressure of the pressure balance, P_{Set} .

The data show, over a period of 24 h, a $\pm 2\sigma$ spread of 0.7 mPa for the empty cavity data (corresponding to spreads in refractivity and beat frequency of 2×10^{-12} and 370 Hz, respectively). For 4303 Pa, the data have a spread of 3 mPa (0.7 ppm) and a mean deviation of -4.7 mPa (1.1 ppm). These spreads are in parity with the best GAMOR data taken with a Zerodur cavity [19]. The difference in spread is attributed mainly to drifts in the temperature assessments and pressure gauge C within the measurement period. The mean deviation between the pressure measured by the refractometer and the set-pressure of the pressure balance at 4303 Pa originates mainly from drifts in the temperature assessments between the instances of characterization and measurements.

Figure 4 displays the Allan deviation of the data presented in Fig. 3 (blue and red markers) together with the best published GAMOR data from a system with a Zerodur spacer (green) [19].

As is expected of GAMOR, which is insensitive to long-term drifts of the cavity length, the Allan deviation of data taken from an empty cavity (in which temperature drifts become irrelevant)

does not show any noticeable drift (blue in Fig. 4). Such measurements are solely limited by white noise, in this case at a level of $3 \text{ mPa s}^{1/2}$, providing a deviation of 0.03 mPa (which corresponds to a deviation of the detected beat frequency of 16 Hz) at 10^4 s . This shows that the system does not pick up any fluctuations or drifts from the cavity within this time period. The data taken at 4303 Pa (red markers) show, for measurement times up to 500 s , a slightly higher white noise level of $7 \text{ mPa s}^{1/2}$, after which disturbances start to affect the system.

This is a clear improvement from the previously used Zerodur cavity for which the white noise levels of the empty cavity measurement and that at 4303 Pa were 10 and $22 \text{ mPa s}^{1/2}$, respectively (where the latter are displayed by the green markers in Fig. 4) [19]. Furthermore, the Allan deviation of the empty cavity measurements in the Invar system was, at longer times, five times better than that of the Zerodur-based system, indicating a reduction in fluctuations or drifts from the cavity [19].

For the present system, the optimum integration time for the 4303 Pa measurement (red in Fig. 4) was found to be around 1000 s (corresponding to 10 gas modulation cycles) at which the Allan deviation has a minimum of 0.34 mPa ($\sim 0.08 \text{ ppm}$). For longer integration times, the deviation increases before it reaches a plateau of 0.7 mPa (at 7000 s). The level of the minimum is significantly better, and that of the plateau is slightly better, than the $0.9\text{--}1 \text{ mPa}$ reached with the Zerodur cavity [19]. The increasing Allan deviation between 1000 and 7000 s is attributed to fluctuations in the temperature measurement module.

In conclusion, this work demonstrates that the GAMOR methodology, with its unique ability to reduce the pick-up of fluctuations and drifts, allows for the use of a wider range of materials as cavity spacers for ultra-sensitive refractometry. It is shown that it is possible to construct a DFPC-based refractometry system based on an Invar spacer that, when used with GAMOR, on time scales between 100 and 10^4 s , outperforms the previously best Zerodur-based system [18,19]. For an empty cavity, the system shows a pure white-noise-limited response up to 10^4 s , at which it provides, for assessment of N_2 , a standard deviation of 0.03 mPa . This indicates that the GAMOR-based system, under these conditions, does not pick up any fluctuations or drifts from the cavity.

Moreover, since deviations between the real and the measured gas temperature affect the assessment of finite pressure, the improved performance at 4303 Pa indicates that the Invar spacer has a more homogeneous temperature distribution than the Zerodur spacer.

It could also be concluded that, on longer time scales (at around 10^4 s), the present realization reaches a stability that is similar to its earlier Zerodur counterpart. This shows that at these time scales, the refractometer is capable of assessing pressure with a precision that is given by the temperature assessment. The system is presently limited by drifts in the temperature measurement module. In a future version of the system, this module will therefore be upgraded, whereby the true long-term properties of the Invar-based GAMOR system can be more clearly assessed.

Finally, taking into account that Invar has a number of advantages with respect to conventionally used spacer materials, and that the system is more compact, has a smaller operating volume, and a lower fabrication cost than previous refractometry

systems, it can be concluded that the Invar system presented here is a clear improvement with respect to a similar Zerodur-based system. This shows that the GAMOR methodology allows for the realization of refractometry systems based on alternative spacer materials that up to now could not be used due to their large thermal expansion.

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