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Deliverable D2: ‘*Report evaluating the performance of the different types of FP-based refractometers developed with respect to their precision, accuracy, working range and target relative uncertainties of 500 ppm in the range 1 Pa – 1 kPa and 10 ppm in the range 1 kPa – 100 kPa* ‘

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Report evaluating the performance of the different types of FP-based refractometers developed with respect to their precision, accuracy, working range and target relative uncertainties of 500 ppm in the range 1 Pa – 1 kPa and 10 ppm in the range 1 kPa – 100 kPa

Deliverable D2 in the EMPIR project 18SIB04, "QuantumPascal"

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Abstract This report summarize performance of the Fabry-Perot refractometers developed in work package 1 in the 18SIB04 QuantumPascal project. The report first briefly introduces the concept of performance in terms of precision, accuracy, and working range. Thereafter each of the systems that have been developed by the partners is shortly described and their contribution to the overall performance in terms of uncertainty is highlighted. From the combined results of the systems, it can be concluded that the target uncertainties have been reached.

1 Introduction

This report, which constitutes Deliverable D2 in the EMPIR project 18SIB04 "QuantumPascal", summarizes the performance of the Fabry-Perot (FP) based refractometers that have been developed within work package 1 (WP1). The work package was setup to target the following overall objective:

- To improve the accuracy and extend the working range of Fabry Perot refractometry quantum-based methods that have the potential to become primary standards of the SI unit of pressure, the pascal. The target uncertainties (k=1) and pressure ranges are 500 ppm in the range 1 Pa - 1 kPa and 10 ppm in the range 1 kPa - 100 kPa"

To reach the target uncertainties, several key issues have been addressed within WP1, such as cavity deformation, the assessment of gas temperature, and gas permeation in the cavity material. While deliverable D1 of 18SIB04, i.e. "*Fabry-Perot-cavity-based refractometry — A guide to its realization and implementation*" [1], describes this work in detail, this report summarizes the overall results in terms of the performance of the systems in relation to the target uncertainties given above.

The report is structured in the following way. First, a short introduction to the various performance concepts, such as precision, accuracy, and working range is given. Thereafter follows a summarizing comment on each of the developed systems, including the evaluated performance when applicable. Finally, the overall results are provided in relation to the target uncertainties.

2 Definition of performance of the FP-based refractometers

To benchmark the performance of the FP-systems developed within WP1, the concepts of precision, accuracy, and working range, are used.

In short, the precision refers to the systems ability to provide results with a given spread in the data. For FP-based refractometers, there are several entities that affect the precision. Most notable; the mechanical stability, the laser-locking scheme, and the efficiency of the measurement procedure. The latter is often of importance as precision is affected by varies types of drifts depending on what timescale the precision is given for. Although precision is important for many application by itself, a high precision is often a prerequisite for a high accuracy (i.e. a low uncertainty).

The accuracy refers to the ability of the systems to produce results with a minimum uncertainty, that is, the closeness of the results to the "true value". For FP-based refractometers there are several entities that affect the accuracy, most notable the uncertainties in the knowledge of gas parameters, the gas temperature, the pressure-induced cavity deformation, outgassing and leaks, as well as the general design of the system.

Finally, an important factor of any given technique is its working range. For FP-based refractometers the working range is in general limited on the lower pressure side by the resolution and on the high side by the mechanical ability to withstand the pressure level.

3 Report on FP refractometers developed within the project

Within the 18SIB04 QuantumPascal, several different partners have developed FP refractometers. Here a summery of these systems and, when applicable, their contribution to the overall target uncertainties of the project are summarized.

3.1 Report on the stationary Invar-based FP refractometer in Sweden

The stationary Invar-based FP-refractometer (in this report referred to as the stationary optical pascal, the SOP), have been described in detailed in a series of papers, see for example [2, 3] and reference therein. The system is also described in some detail in the associated guide "*Fabry-Perot-cavity-based refractometry — A guide to its realization and implementation*" [1].

In short, the SOP is based on a dual FP-cavity realized in an Invar spacer that operates using narrow linewidth fiber lasers at 1550 nm. To asses the gas temperature, the cavity is directly coupled to an external gallium fixed-point cell that operates intermittently (in cycles of a number of days) during the measurements, providing the system with the ability to measure the gas temperature with an (k=2) uncertainty at the 5 ppm-level [4]. The system utilizes the GAMOR methodology, which, by modulating the gas in the cavity, makes the system resilient against linear drifts and fluctuations, which, in turn, improves on its precision [5, 6].

3.1.1 Evaluated precision

To evaluate the precision of the system, two different studies were performed. The first concerned the precision when assessing a certain set pressure. It was concluded by Silander et al. [2], when assessing the pressure of an empty cavity (continuously evacuated) over 24 hours, that the system had a (k=2) repeatability of 0.7 mPa. When assessing pressure set by a pressure balance (4303 Pa), it demonstrated that a (k=2) precision of 3 mPa, corresponding to 0.7 ppm could be achieved. The decrease in precision, as compared to the empty cavity measurement, was mainly attributed to drifts in the temperature assessment.

In the second study, the short-term precision (typical providing assessments with sub-second resolution for periods up to hundreds of seconds) was assessed by connecting the SOP and the similarly constructed transportable Invar-based FP refractometer (also referred to as the TOP, see below), simultaneously to a pressure balance [7]. By operating both the TOP and SOP independently, and having them to assess the same pressure set by the pressure balance, it could be shown that the systems achieved a joint short-term recision at the 0.04 ppm-level.

3.1.2 Evaluated accuracy

To evaluate the accuracy of the SOP, the system was subjected to a careful characterization in Silander et al. [3]. In this work, it was concluded that the SOP system has, from 1 Pa to 25 kPa, an expanded uncertainty (k=2) of $[(10 \text{ mPa})^2 + (10 \times 10^{-6} P)^2]^{1/2}$.

After this characterization, for which the upper range was limited to 25 kPa due to limitation of the vacuum system, the system has been upgraded to allow for pressures up to 100 kPa. Although no full characterization has been performed after the upgrade, it is estimated that the system will retain its performance also in the range 25-100 kPa.

It should be noticed though that, in Silander et al. [2], which addressed an earlier version of the Invar-based refractometry system, the SOP demonstrated a significantly (one order of magnitude) better performance in terms of the empty cavity frequency stability as compared to the aforementioned assessment [3] [0.7 mPa as compared to the 7 mPa (k=2)]. The reason for this deterioration in vacuum pressure uncertainty is present unclear, but will be the subject of an upcoming study.

3.2 Report on the transportable Invar-based FP refractometer in Sweden

The TOP has been described in detailed in several works, see [7–9] and reference therein. The system is also described in the guide "*Fabry-Perot-cavity-based refractometry — A guide to its realization and implementation*" [1].

In short, the system is similarly to the SOP system described in section 3.1. However, it is not equipped with an external gallium fixed-point cell. Instead it is utilizing calibrated Pt-100 sensors for temperature measurements. Its ($k = 2$) uncertainty has been estimated to be 26 ppm.

3.2.1 Evaluated precision

As was described section 3.1.1, the precision of the TOP was investigated by Forssén et al. [7] together with that of the SOP. It was concluded in that work that the system has, when operating using GAMOR-cycles, a short-term precision on the 0.04 ppm-level. In terms of empty cavity repeatability it is estimated to have a similar, but slightly worse (due to mechanical instabilities), performance as the SOP.

3.2.2 Evaluated accuracy

The TOP was carefully characterized together with the SOP in terms of its extended uncertainty by Silander et al. [3]. It was concluded in that work that the TOP system had an extended uncertainty, for pressures in the 1 Pa to 25 kPa range, of $[(16 \text{ mPa})^2 + (28 \times 10^{-6} P)^2]^{1/2}$ (k=2). This results is slightly worse than the SOP (as was described in section 3.1.2). While the difference in the constant term (16 mPa compared to 10 mPa) can be attributed to a slightly less optimized vacuum system and a worse mechanical stability, the difference in the proportional term (28 ppm compared to 10 ppm) can be attributed to the dissimilar means of the temperature assessment.

As with the SOP, also the TOP has, since the common characterization, been upgraded to allow for pressures up to 100 kPa. However, again as for the SOP, no new characterization has been performed after its upgrade.

3.3 Report on Zerodur-based FP refractometer in France

Cnam has within the scope of the project, developed a Zerodur-based FP refractometer for pressure measurements in the1 Pa - 100 kPa pressure range with special emphasis on temperature regulation and assessment. The refractometer is based on a single cavity composed of a cubic spacer of 50 mm edge length in Zerodur® with two optically bonded plano-concave fused silica mirrors. The refractometer operates using two frequency doubled Nd:YAG lasers at 532 nm: one is locked to a transition in iodine, while the other is locked to the FP cavity.

The refractometer temperature is regulated to the gallium melting point with sub-mK stability for days by the use of autonomous systems using Pt-100 sensors and thermofoils (see guide A1.2.3 of this project). The temperature is assessed with a calibrated SPRT (glass-capsuled from Fluke Calibration) Pt-25 placed in a specially drilled hole in the lid of an internal copper chamber that includes the cavity. This sensor is placed as close as possible to the refractometer. By this, we assume that the copper temperature is the same as the gas temperature.

The system has not yet been subject to a detailed uncertainty budget, but some first estimates of the performance are given in Appendix A.

However, within the project, it was shown that before the most recent update of the system, that outgassing and leak rates were below the limit corresponding to the target vacuum pressure uncertainty (500 ppm at 1 Pa) [10].

3.4 Report on the dichroic FP refractometer in Germany

At PTB, work begun with the realization of a sapphire-based system. Such a system holds a lot of potential as it has a very low pressure-induced deformation compared to other systems, as well as virtually no gas permeation. It also has the possibility to act as a thermometer in itself, as is described in some detail in "*Fabry-Perot-cavity-based refractometry — A guide to its realization and implementation*" [1]. However, as a sapphire-based system also is highly complex and difficult to manufacture, the undertaking of such a system became exceedingly time-consuming. Therefore, various FPC-based systems using Zerodur or Invar spacers, operating at both 633 nm and 1550 nm, have meanwhile been investigated. These undertakings have improved the understanding of possible development potentials with regard to the planned sapphire-based and other systems. Although promising partial results have been achieved during the project, no complete uncertainty budget of any sapphire-based system has yet been performed.

3.5 Report on the NEXCERA-based FP refractometer in Spain

CEM has developed a dual FP cavity refractometer, based on a NEXCERA cylindrical spacer and two ClearCeram Z (CCZ) mirrors for the 1 Pa to 100 kPa pressure range, operating at 633 nm. NEXCERA CD107 was chosen as material with the motivation that it has a low susceptible to pressure-induced deformations due to its high stiffness.

The system is temperature stabilized by three temperature control systems: The exterior system controls the temperature to 100 mK, the intermediate system controls the temperature within 10 mK and the most interior system, which holds the cavities, controls the temperature to to 1 mK.

The gas temperature is measured by two Pt-25 probes for each cavity, one inside the copper sleeve while the another one is in contact with the gas.

The system is still under development and have not yet undergone any uncertainty evaluation.

4 Conclusion

In conclusion, several instruments have been developed within the project in order to reach the target uncertainties. While only a couple of systems have undergone proper uncertainty evaluation in terms of their ability to assess pressure, the joint efforts and achievements clearly shows that the target uncertainties not only can be reached, by also exceeded if the individual realizations are combined.

Figure 1 provides a summary of the results of the endeavours of the consortium. The dashed straight black lines indicate the target uncertainties (500 ppm from 1 Pa to 1 kPa and 10 ppm from 1 kPa to 100 kPa). The blue and the orange thick curves show the evaluated uncertainties of two independent system, the SOP and the TOP systems, respectively [3], while the purple dashed curve shows the performance of a hypothetically system based on the combined best findings of various systems within the project.

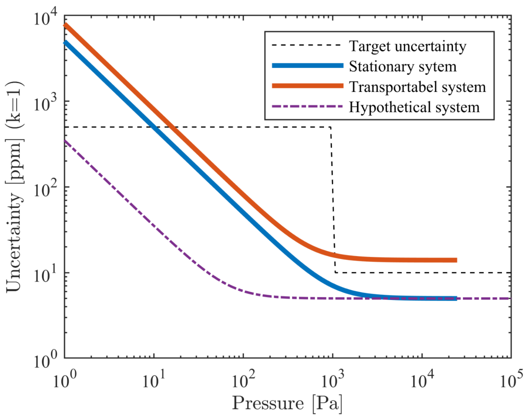

Figure 1. Performance of the FP-based refractometers developed within this project. The black dashed lines correspond to the target uncertainties of the project. The blue and orange lines correspond represent the performance of the SOP and TOP systems that were evaluated in [3]. The purple dashed-doted line represents the performance of a hypothetical, but not yet realized system, which is based on the combined best findings of various systems within the project.

From the figure it can be noted that in the range from 10 Pa to 25 kPa, the SOP system was evaluated to have a lower uncertainty than the target benchmarks. However in the range below 10 Pa, the system exhibited, at the time of the characterization, limitations due to its empty cavity repeatability, outgassing and leaks, and the ability to estimated the residual pressure. Furthermore, the system was not explicitly characterized above 25 kPa due to, again at the time of the characterization, limitations in the vacuum system.

To demonstrate the readiness of the developed technology within the project, the figure also indicates the performance of a hypothetical, but fully realistic, system based on the most prominent achievements within the project. While the realized systems were limited due to the factors given above, the hypothetical system takes into account the demonstration of the much lower empty cavity repeatability from [2], the low out gassing and leak rate of the Cnam-system [10], and the inclusion of a significant, and straightforwardly implemented, means to assess the residual pressure. Furthermore, for pressures above 25 kPa, the hypothetical system includes the estimated capability of the systems after the upgrades of their vacuum systems.

To conclude, the target uncertainties have been reached and demonstrated in most part of the working range. In the parts where the target uncertainties was not explicitly reached by a single operating system, it has, within the project, been shown to be well within the reach of the technology if the joint achievements of the developed systems are combined.

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Appendix

A. Detailed description of the Cnam system

Cnam has within the scope of the project developed a Zerodur-based FP refractometer for pressure measurements in the 1 Pa - 100 kPa range with special emphasis on temperature regulation and assessment. The refractometer is based on a single cavity composed of a cubic spacer of 50 mm edge length in Zerodur® with two optically bonded plano-concave fused silica mirrors. The refractometer operates using two frequency doubled Nd:YAG lasers at 532 nm: one is locked to a transition in iodine, while the other is locked to the FP cavity. This allows for good assessment of the cavity mode frequency.

This refractometer configuration has the advantage of simplifying the design of the cavity, in particular regarding the integration of the temperature sensor, which in placed in the center of the refractometer. On the other hand, because of the use of the single cavity, the system can be sensitive to long term-drift.

In addition, in order to reach the uncertainty target, the system must be temperature regulated and assessed to the mK scale. For that, the temperature is regulated to gallium melting point with sub-mK stability, which can be achived for days using by autonomous systems using Pt-100 sensors and thermofoils (as is described in some detail in the A1.2.3 guide of this project). The temperature is assessed with a calibrated Pt-25 SPRT (glass-capsuled from Fluke Calibration) placed in a specially drilled hole in the lid of an internal copper chamber that includes the cavity. This sensor is placed as close as possible to the refractometer (about 4 mm from it) and we assume that the copper temperature is the gas temperature. The uncertainty after calibration was assessed to 0.7 mK.

Characterisation of the refractometer To measure the pressure, in addition to measuring the laser frequency when the refractometer is in vacuum (which can be done with a stability of around 100 Hz, which corresponds to *ca.* 0.1 mPa) and in gas (which can be assessed at 100 kPa with a 10 kHz stability, which corresponds to *ca.* 7 mPa), the intrinsic parameters of the refractometer must be well defined with their corresponding uncertainties. In this way, the refractometer has been fully characterised by measuring the coefficient of thermal expansion (CTE), the Free Spectral Range (FSR), the pressure-induced distortion coefficient and the long-term drift (ageing).

It is not yet possible to propose a detailed uncertainty budget though. A first estimation of the uncertainty contribution of the intrinsic parameters is given as follows:

- **Ageing**: The long-term drift of the refractometer was estimated to $2.129(14) \times 10^{-10}$ day^{-1}. The correction is about 3 mPa with an uncertainty << 1 mPa if we consider one hour between gas measurement and reference state in vacuum ;
- **CTE**: The measured value of the CTE is $1.58(12) \times 10^{-7}$ K^{-1}. This corresponds to an uncertainty contribution of about 3 mPa for a 1 mK uncertainty of the temperature variation between reference and measurement state. This contribution (0.04 ppm) can be lowered by a factor 10 with a lower CTE. This can be achieved by changing the material of the mirrors to a CTE with a value closer to that of the best Zerodur ones (1.6×10^{-8} K^{-1}) ;
- **FSR** : The measured value is 2997812.6(1.8) kHz and the uncertainty contribution is about 5×10^{-7} (*i.e.* 0.5 ppm);
- **Pressure-induced distortion coefficient**: This coefficient has been estimated both theoretically by simulation and experimentally ($6.70(2) \times 10^{-12}$ Pa^{-1}). The relative uncertainty contribution for pressure measurements with nitrogen is 6 ppm. Future works are planned to check this experimental value using the two-gas method and reassess the A_R molar polarizability for nitrogen and argon at 532 nm with a targeted relative uncertainty $< 5 \times 10^{-5}$.