Measurement: Sensors 18 (2021) 100253

Contents lists available at ScienceDirect

Measurement: Sensors

journal homepage: www.sciencedirect.com/journal/measurement-sensors

Rayleigh scattering for pressure assessment

ARTICLE INFO

Keywords
SI
Pascal realization
Optical pressure standard
Rayleigh scattering
Pressure metrology

ABSTRACT

This paper reports about the realization of a pioneering system for pressure measurements, currently tested up to 400 kPa, based on the Rayleigh scattering and with no mechanical parts interacting with the gas. The preliminary results showed that the device exhibits a strong linear dependence between gas pressure and scattered light, of the order of 10^{-5}, paving the way for promising innovative realizations of scattering-based system for pressure measurement with a single and simple device in a wide pressure range between 10 Pa and 1 MPa.

1. Introduction

Present realizations of the pascal are based on mechanical devices as pressure balances [1–3] and mercury manometers [4] or systems operating through static or dynamic expansion of pure gas [5–8]. Such realizations are subject to inherent limitations because of safety (recent Word Health Organization resolutions recommending the progressive reduction of human exposure to mercury and mercury compounds) and practical reasons (bulkiness, fragility, complexity of operation).

The 26th General Conference on Weights and Measures (November 16, 2018) adopted a revision of the International System of Units (SI). The new SI, in effect since May 2019 [9], is based on established values of several fundamental physical constants and encourages the exploration of new paths for vacuum and pressure measurements, starting by the fundamental laws and constants of physics. Within this frame-work, the new trend is to measure directly the gas density instead of traditional force per unit area, by means of devices based on light-matter interaction [10]. In recent years, several studies have been performed to assess optical methods as new route for pressure measurements, through the use of Lorentz-Lorenz equation [11–19].

This paper reports the realization of an innovative system for pressure measurements based on Rayleigh scattering, currently tested up to 400 kPa. The preliminary results showed that system can provide a large dynamic range with excellent linearity: as the theory of Rayleigh scattering proves a direct dependence of scattered light on molar polarizability, a new version of Rayleigh scattering-based system for pressure assessment will be realized and will be also used as a comparator instrument to measure the polarizability of different gases, starting from helium as the reference gas, whose polarizability can be determined through ab-initio calculations with high accuracy [20,21].

2. Rayleigh scattering

The phenomenon of light scattering is a typical manifestation of light-matter interaction. A strong contribution to the study of this fascinating phenomenon was made between the second half of the 19th century and the beginning of the following century, allowing to answer ancient questions regarding the origin of the colour of the sky and the sunset [22–27].

The Rayleigh scattering, in particular, refers to elastic scattering of light from particle having small diameter comparing to the wavelength of the incident light; it is named after John William Strutt (Lord Rayleigh), a scientist who gave a huge contribution to the advancements of physics in different fields and received the Nobel Prize for Physics in 1904 “for his investigations of the densities of the most important gases and for his discovery of argon in connection with these studies".

The classical description of Rayleigh scattering is related to the effect of the incident radiation on a small particle: the electrons in the atoms are induced to oscillate by the applied electromagnetic field and irradiate, like a dipole antenna. This simple model provides an excellent explanation for spherical molecules such as argon and helium: in this case, for molecules having a spherical symmetry, the induced dipole moment vector $\boldsymbol{p}$ is in the same direction of the polarization of incident light. The linear relation between the dipole moment and the incident electric field $\boldsymbol{E}_\mathbf{i}$ is defined through a scalar coefficient, the polarizability α, according to:

$$\boldsymbol{p} = \alpha \boldsymbol{E}_i \tag{1}$$

Let us consider the case of an electromagnetic radiation propagating along the x-axis direction and linearly polarized along z-axis (Fig. 1). Under the previous assumptions of small and spherical molecules, the intensity of scattered light I_s from a single molecule, at the distance $\boldsymbol{r}$, is given by Ref. [28]:

$$I_s = I_i \frac{\pi^2 \alpha^2}{\varepsilon_0^2 \lambda^4 r^2} sin^2(\phi) \tag{2}$$

where I_i is the intensity of incident light, ε_0 the vacuum permittivity, λ the vacuum wavelength and ϕ is the angle at which the scattering light is observed, i.e. the angle projection of the dipole moment vector on the observation direction. Thus, the dependence of light scattering by λ^4 gives us an immediate explanation of the colour of the sky, as shorter wavelength like blue, are more strongly scattered than longer wavelength. It should also be noted that the scattered intensity is maximum at

https://doi.org/10.1016/j.measen.2021.100253

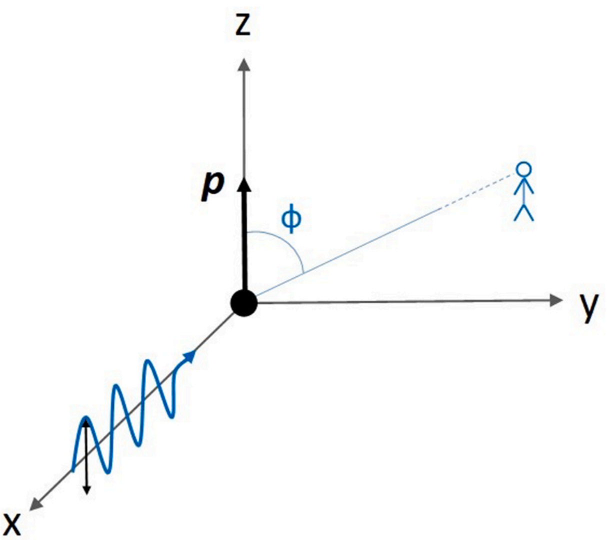

Fig. 1. Schematic of scattered light geometry for small and spherical molecule.

an angle $\phi = \pi/2$ and evidences a quadratic dependence on the polarizability α.

Rayleigh scattering, as well as the Mie scattering for spherical molecules of similar size to the wavelength, can be considered elastic processes in which the total kinetic energy is conserved and the wavelength and the frequency of the radiation don't change: this peculiarity makes easier the definition of the "dynamic" polarizability, to be intended as the mean dynamic polarizability at the wavelength λ. Furthermore, considering a volume of interaction between incident light and gas molecules, the intensity of scattered light is proportional to the number of molecules in the volume of interaction, namely depends on the number density [29]; according to gas equations of state, the number density is proportional to temperature and pressure, therefore, at the same temperature, the gas pressure is linearly proportional to the number of scattered photons.

3. Experimental set-up and results

The simplified scheme of the experimental set-up is shown in Fig. 2: a short wave (blue) CW laser interacts with the gas in a chamber. As stated previously, because of the Rayleigh scattering, each gas atom or molecule immersed in the electromagnetic field acts as a dipole oscillating at the EM frequency and irradiating energy proportionally to the gas polarizability. A photon collecting system allows to accurately measure the strength of the interaction, which depends on the gas pressure.

The realized apparatus (Fig. 3) is essentially formed by a stainless steel cross-shaped chamber equipped with two optical viewports, respectively for the entrance of laser light and the exit of scattered light towards the detector. In this preliminary realization, invented by M. Pisani [30], the cross-shaped chamber has been realized simply using ISO-KF vacuum flanges with metal gasket; the light source is a blue laser at 460 nm, having a power of 500 mW. A series of baffles and absorbers have been used to reduce the effect of spurious light on the signal collected by the detector. The scattered light is collected by a system, whose main component is the camera Ascent A4000, a cooled, 16 bit, 4 Mpixel CCD camera, equipped with a monochrome sensor and with dimension of each pixel equal to 7.4 μm × 7.4 μm. In the opposite side of the laser entrance, a 90° pipe bend system has been added to limit backwards reflection of light and to connect the gas inlet system and the oil-free vacuum pump system.

The whole system has been mounted on an aluminium breadboard (450 mm × 450 mm), so that it was easily transportable. The first realization described in this paper is not equipped with any temperature measurement and control and no pressure regulation: this "as is" set-up has been essentially realized to test the performance of the Rayleigh scattering-based system in term of linearity between the scattered light intensity and gas pressure, to evaluate the potentiality of this innovative technique for gas pressure assessment.

A series of measurement has been carried out with different gases, i. e. nitrogen, argon and helium: at each pressure points measured by a Mensor CPG2500 instrument, the correspondent image of scattered light detected by the CCD camera has been acquired up to about 400 kPa (in case of nitrogen).

A custom software in python™ ambient has been developed to analyse the acquired images (Fig. 4), with the double aim of detecting and correct the effect due to eventual spurious scattering centres and calculating the light intensity using two different techniques: an integral sum or a Gaussian fit.

After having acquired the camera images at each pressure value from the experiment, with the appropriate acquisition parameters, data are analysed by the software. Each dataset for a given gas molecule or

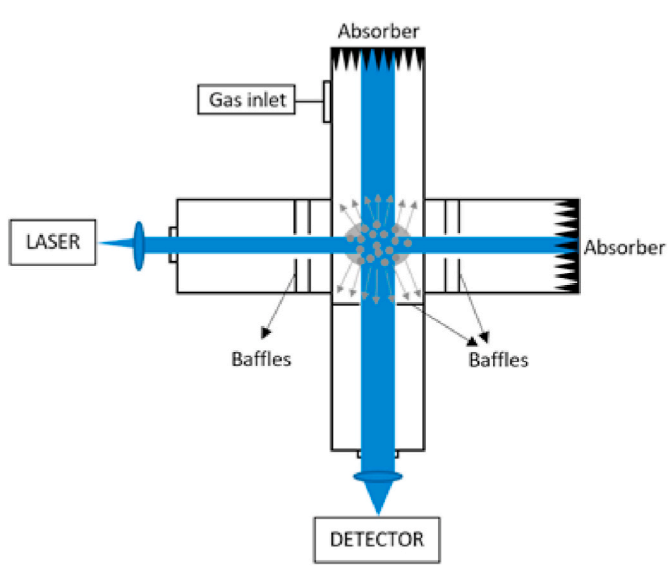

Fig. 2. Scheme of the experimental set-up.

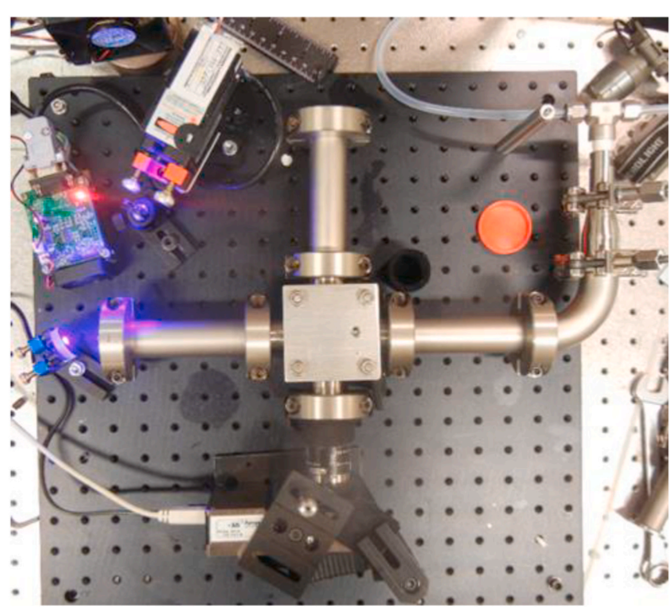

Fig. 3. Picture of the practical preliminary realization.

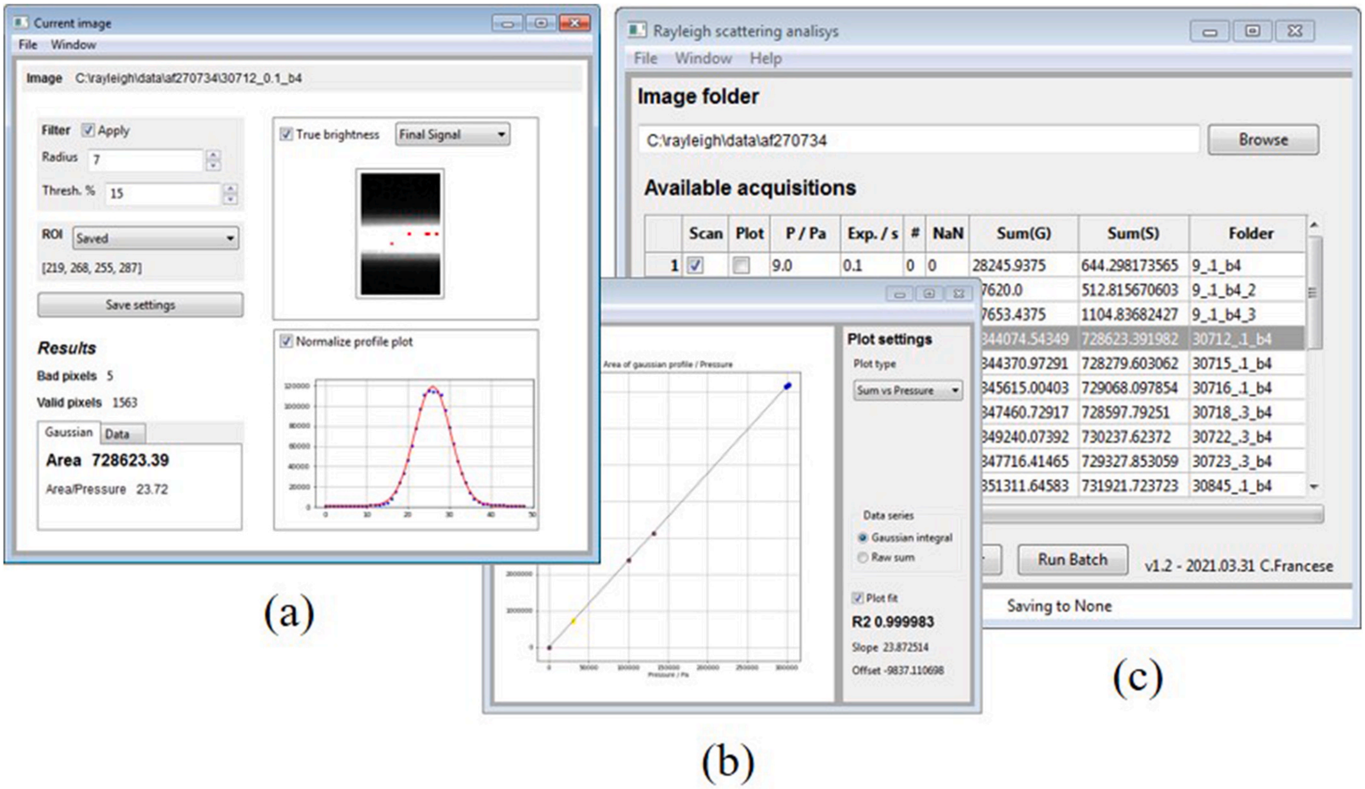

Fig. 4. Software for the analysis of image data of pressure for helium (Gaussian fit).

experimental conditions corresponds to a single folder, with a strict naming convention for each file including the pressure value and acquisition parameters, thus allowing to easily process all the images in one shot (Fig. 4c). Before starting the dataset analysis, each image can be visually checked for possible acquisition issues, i.e. perturbation of the scattered light due to spurious particles in the beam –red dots in Fig. 4a – and the beam profile is shown. The red curve reported in the same figure is estimated beam profile calculated by fitting the series p_x of the average values of the image along each column $p_x = \frac{1}{N_y}\sum_{1}^{N_y} p_{x,y}$ with a Gaussian function where $p_{x,y}$ is the raw value of each pixel of the image.

As the time of writing, two methods have been tested for the intensity estimation of the total scattered light at each pressure level: the first consists in the raw sum of each pixel value and the second method analytically integrates the Gaussian profile fitted data. Tens of runs of the software using both methods on different datasets showed that the values of coefficient of determination are pretty the same. The resulting curve of the scattered light intensity vs. pressure value is shown in Fig. 4b and the relevant linear fit results are reported.

Figs. 5–7 show the results for nitrogen, argon and helium, in which the scattered light intensity, at different nominal pressures, has been calculated by a Gaussian fit.

The realized preliminary set-up has highlighted encouraging results, showing an excellent linearity of the system and opening the way to a new improved realization of a Rayleigh scattering-based device for gas pressure measurement.

4. Toward a novel version of Rayleigh scattering-based system (RAY)

A novel version of the Rayleigh scattering-based system, named RAY, is under development in the framework of the 18SIB04 QuantumPascal EMPIR Project [31].

The novel experimental set-up will benefit from a new design, which

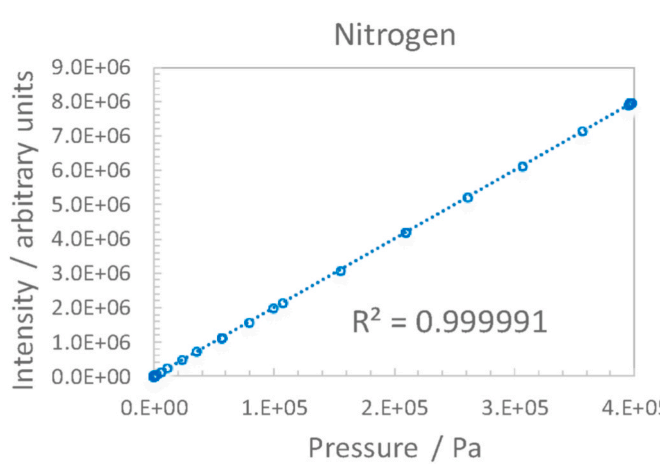

Fig. 5. Scattered light intensity as a function of pressure for nitrogen (Gaussian fit).

mainly aims to introduce and optimize temperature measurement and control, minimize the stray light phenomenon and realize a unique device able to work over a wide pressure range from 10 Pa to 1 MPa.

In the new layout, as shown in Fig. 8, the cross-shaped vacuum chamber is directly dug in an aluminium block (210 mm × 210 mm × 70 mm) and equipped with five ConFlat® ports, to connect four optical viewports and a gas inlet and pumping system. The custom viewports have been designed and realized to work up to an over-pressure of 2.5 MPa, which is conservative, considering that the expected upper limit of RAY is 1 MPa. The chamber may host until six temperature sensors to implement the temperature measurement and control and it will be equipped with special absorbers, able to decrease the stray light effect of a factor 10^{-5}.

The Rayleigh-based technique is expected to provide a large dynamic

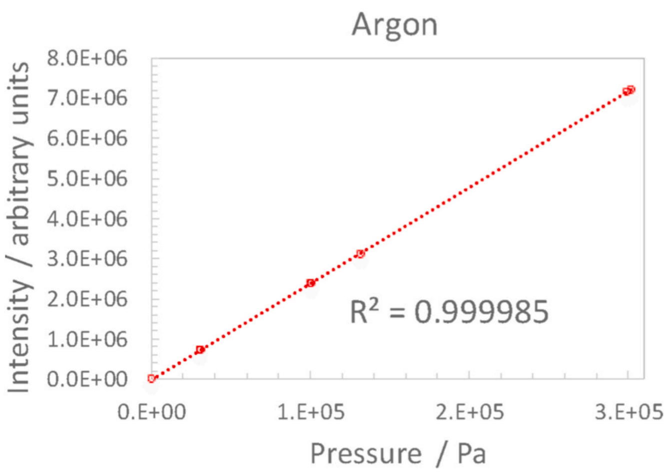

Fig. 6. Scattered light intensity as a function of pressure for argon (Gaussian fit).

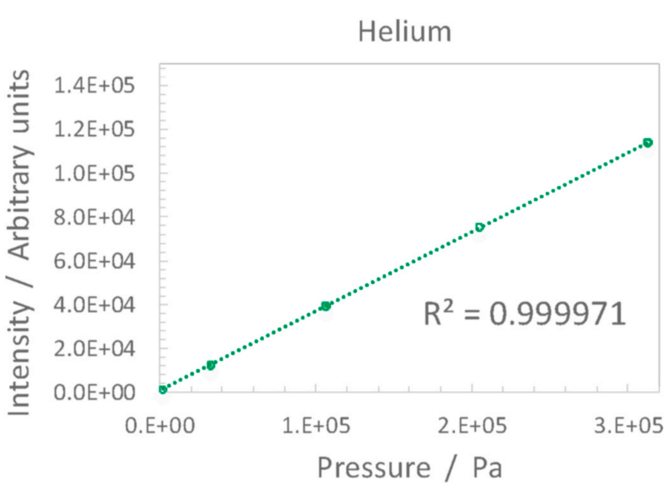

Fig. 7. Scattered light intensity as a function of pressure for helium (Gaussian fit).

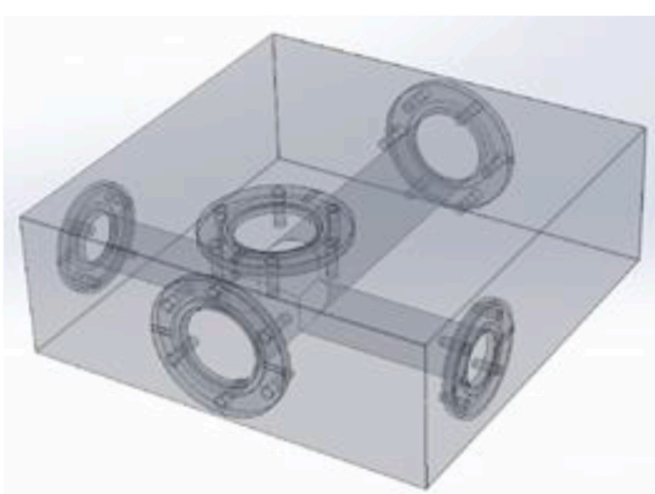

Fig. 8. Aluminium vacuum chamber for RAY.

range with excellent linearity. In addition, the theory of Rayleigh scattering proves a direct dependence of scattered light on polarizability (eq. (2)). As consequence, the novel realization, RAY, could be also used as a comparator instrument to measure the polarizability of different gases, starting from helium as the reference gas, whose polarizability can be determined through ab-initio calculations.

5. Conclusions

An innovative system for pressure assessment has been realized at INRiM. It is based on the measurement of the Rayleigh scattering of gas molecules. A preliminary experimental set-up has been tested up to 400 kPa, in terms of linear response between scattered light intensity and gas pressure, exhibiting a linearity of the order of 10^{-5} and paving the way to a new improved realization.

A novel version is under development in the framework of the 18SIB04 QuantumPascal EMPIR Project. The new design aims to introduce and optimize the temperature measurement and its related active control, minimize the stray light phenomenon, which is a significant limiting factor of accuracy of the system, and realize a unique device able to work over a wide pressure range from 10 Pa to 1 MPa.

Acknowledgments

This work was, in part, supported by Quantum Pascal project (18SIB04), funded through the European Metrology Programme for Innovation and Research (EMPIR). The EMPIR initiative is co-founded by the European Union's Horizon 2020 research and innovation program and the EMPIR Participating States.

References

[1] C.G. Rendle, H. Rosenberg, New absolute pressure standard in the range 1 Pa to 7 kPa, Metrologia 36 (1999) 613–616.
[2] A. Ooiwa, A heterodyne laser interferometric oil manometer, Metrologia 30 (1994) 607–610.
[3] C.M. Sutton, M.P. Fitzgerald, Performance aspects of gas-operated pressure balances as pressure standards, Metrologia 46 (2009) 655–660.
[4] F. Alasia, G. Birello, A. Capelli, G. Cignolo, M. Sardi, The HG5 laser interferometer mercury manometer of the IMGC, Metrologia 36 (1999) 499–503.
[5] W. Jitschin, High-accuracy calibration in the vacuum range 0.3 Pa to 4000 Pa using the primary standard of static gas expansion, Metrologia 39 (2002) 249–261.
[6] M. Bergoglio, A. Calcatelli, Uncertainty evaluation of the IMGC-CNR static expansion system, Metrologia 41 (2004) 278–284.
[7] K. Jousten, H. Menzer, D. Wandrey, R. Niepraschk, New, fully automated, primary standard for generating vacuum pressures between 10^{-10} Pa and 3×10^{-2} Pa with respect to residual pressure, Metrologia 36 (1999) 493–497.
[8] M. Bergoglio, D. Mari, INRIM continuous expansion system as high vacuum primary standard for gas pressure measurements below 9×10^{-2} Pa, Vacuum 84 (2009) 270–273.
[9] Bureau International des Poids et Mesures, The international system of units (S) 9th edition, available online at, https://www.bipm.org/en/publications/si-brochure, 2019.
[10] K. Jousten, J. Hendricks, D. Barker, K. Douglas, S. Eckel, P. Egan, J. Fedchak, J. Flügge, C. Gaiser, D. Olson, J. Ricker, T. Rubin, W. Sabuga, J. Scherschligt, R. Schödel, U. Sterr, J. Stone, G. Strouse, Perspectives for a new realization of the pascal by optical methods, Metrologia 54 (2017) S146.
[11] L.R. Pendrill, Refractometry and gas density, Metrologia 41 (2004) S40.
[12] J. Ricker, J. Hendricks, P. Egan, J. Stone, K. Douglass, G. Scace, Towards photonic based pascal realization as a primary pressure standard, J. Phys. Conf. 1065 (2018) 162018.
[13] P. Egan, J. Stone, J. Hendricks, J. Ricker, G. Scace, G. Strouse, "Performance of a dual Fabry–Perot cavity refractometer", Opt Lett. 40 (2015) 3945.
[14] I. Silander, T. Hausmaninger, C. Forssen, M. Zelan, O. Axner, Gas equilibration gas modulation refractometry for assessment of pressure with sub-ppm precision, J. Vac. Sci. Technol. B 37 (2019), 042901.
[15] I. Silander, C. Forssen, J. Zakrisson, M. Zelan, O. Axner, Invar-based refractometer for pressure assessments, Opt Lett. 9 (2020) 2652–2655, 45.
[16] D. Mari, M. Pisani, M. Zucco, An optical interferometer for pressure measurement, J. Phys. Conf. 1065 (2018) 162007.
[17] D. Mari, M. Pisani, M. Zucco, Towards the realization of an optical pressure standard, Measurement 132 (2019) 402–407.
[18] P. Rourke, C. Gaiser, B. Gao, D. Madonna Ripa, M.R. Moldover, L. Pitre, R. J. Underwood, Refractive-index gas thermometry, Metrologia 56 (2019), 032001.
[19] Z. Silvestri, D. Bentouati, P. Otal, J.-P. Wallerand, Towards an improved helium-based refractometer for pressure measurement, ACTA IMEKO 9 (Number 5) (December 2020) 305–309.
[20] J.W. Schmidt, R.M. Gavioso, E.F. May, M.R. Moldover, Polarizability of helium and gas metrology, Phys. Rev. Lett. 98 (2007) 254504.

[21] C. Gaiser, B. Fellmuth, Polarizability of helium, neon and argon: new perspectives for gas metrology, Phys. Rev. Lett. 120 (2018) 123203.
[22] J. Tindall, On the action of rays of high refrangibility upon gaseous matter, Trans. Roy. Soc. London 60 (1870) 333–365.
[23] J.W. Strutt, Lord Rayleigh, On the light from the sky, its polarization and colour, Philos. Mag. Ser. 4 (41) (1871) 107–120.
[24] J.W. Strutt, Lord Rayleigh, On the scattering of light by small particles, Philos. Mag. Ser. 4 (41) (1871) 447–454.
[25] G. Mie, Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen, Ann. Phys. 330 (3) (1908) 377–445.
[26] J. Cabannes, “Sur la diffusion de la lumière par l'air”, C. R. Acad. Sci. 160 (1915) 62–63.
[27] C.V. Raman, A new radiation, Indian J. Phys. 2 (1928) 387–398.
[28] R.B. Miles, W.R. Lempert, J.N. Forkey, Laser Rayleigh scattering, Meas. Sci. Technol. 12 (2001) R33–R51.
[29] W.L. Eberhard, Correct equations and common approximations for calculating Rayleigh scatter in pure gases and mixtures and evaluation of differences, Appl. Opt. 49 (7/1) (2010) 1116–1130.
[30] M. Pisani, System for Determining the Characteristics of a Gas and Related Method for Measuring Such Characteristics, *Patent EP3394595 B1*, Proprietor: INRiM, Granted 11 March 2020, 2020.
[31] Euramet Project 18SIB04 (QuantumPascal) Towards Quantum Based Realisations of the Pascal, 2019-2022. https://www.ptb.de/empir2019/quantumpascal.

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