Supplemental Document

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Gouy phase in the presence of gas in Fabry-Perot refractometers: supplement

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1. PROPERTIES OF A FREELY PROPAGATING GAUSSIAN BEAM

A. Definitions

A Gaussian beam is an idealized description of electromagnetic radiation whose cross-sectional envelope (its electrical field strength as well as its irradiance) has a Gaussian shaped profile. The mathematical description of such a beam is the solution to the paraxial wave equation [1] (or the paraxial Helmholtz equation [2]) [3] and, as is given both in textbooks and in the literature, can, when propagating in the z direction, be written as [1, 4–7]

$$E(x,y,z,t) = E_0 \frac{w_0}{w(z)} e^{-\frac{(x^2+y^2)}{w^2(z)}} e^{ikz} e^{i\frac{k(x^2+y^2)}{2R(z)}} e^{-i\psi(z)} e^{-i\omega t}, \tag{S1}$$

where E_0 is the electric field amplitude at the origin (i.e. $E(0,0,0,0)$), w_0 is the semi-diameter of the beam waist at the plane where the phase front is flat (i.e. at $z=0$), $w(z)$ is the beam waist at the position z, k is the wave number (given in radians per meter by $\frac{2\pi}{\lambda} = \frac{n2\pi}{\lambda_0}$ where λ is the wavelength in the medium in which the beam is propagating, n is the index of refraction of the medium in which the beam propagates, and λ_0 is the wavelength in vacuum), $R(z)$ is the radius of curvature of the beam's wavefront (at position z), $\psi(z)$ is the Gouy phase (likewise at position z), and ω is the angular frequency of the light (given by $2\pi\nu$, where ν is the frequency of the light). The z parameter represents the distance along the beam counted from the focal plane.

Of these, the semi-diameter of the beam waist at the position z, $w(z)$, is given by

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2}, \tag{S2}$$

the radius of curvature of the wavefront, $R(z)$, can be expressed as

$$R(z) = z\left[1 + \left(\frac{z_R}{z}\right)^2\right], \tag{S3}$$

while the Gouy phase is given by

$$\psi(z) = \arctan\left(\frac{z}{z_R}\right), \tag{S4}$$

where, in all cases, z_R is the Rayleigh range (also referred to as the Rayleigh length in the literature), which is given by

$$z_R = \frac{\pi w_0^2}{\lambda} = \frac{n\pi w_0^2}{\lambda_0}. \tag{S5}$$

In addition, the divergence angle in the far field of a Gaussian beam, θ, is given by

$$\theta = \frac{\lambda}{\pi w_0} = \frac{\lambda_0}{n\pi w_0}. \tag{S6}$$

Equation (S1) provides, together with its associated expressions, i.e. the Eqs. (S2) - (S6), an adequate description of the propagation of beam-shaped light with a Gaussian intensity distribution in a homogeneous medium, i.e. both in vacuum and in a medium with an index of refraction of n.

B. Do the Gouy phase and the Rayleigh range of a freely propagating Gaussian beam have any dependence on the gas?

Equation (S4) shows that the Gouy phase of a freely propagating Gaussian beam [8] depends on the Rayleigh range, which, in turn, according to Eq. (S5), can be interpreted as the ratio of the product of the index of refraction, n, and the area of the beam in its waist, $\pi\omega_0^2$, and the vacuum wavelength of the light, λ_0. This shows that the Rayleigh range does not only have an explicit dependence on the index of refraction, it might also have an implicit one through the beam waist which might be affected by the presence of the gas.

However, it is not clear from the Eqs. (S1) - (S5) to which extent a freely propagating Gaussian beam will be affected by the presence of gas with a given refractivity. The reason for this is that a Gaussian shaped laser beam is characterized (and described) not only by its wavelength, λ (or λ_0), and the electric field amplitude at the origin, E_0, [9], it is additionally characterized by either the semi-diameter of the beam waist, w_0, or the Rayleigh range, z_R, of which none is uniquely defined solely in terms of the entities above; they are only defined when either of them, or the divergence angle, θ, is additionally specified.

2. RAYLEIGH RANGE AND SINGLE-PASS GOUY PHASE FOR A CONCAVE-CONCAVE AND A PLANO-CONCAVE CAVITY

The locking of a laser field to a mode of the cavity sets up constraints that affects the properties of the light.

One of these is the phase resonance condition, which, in the presence of the phenomena that is considered to affect the system, determines the frequency of the light. For the case when the heating of the mirrors by the laser light is neglected and for working ranges centered on the mirror center frequency, but in the presence of the pressure-dependent component of the Gouy phase discussed in this work, it is given by Eq. (14) in the main document.

In addition to this, the light is affected by the constraint that the radii of curvature of the wavefronts of the light at the positions of the mirrors need to be equal to those of the mirrors [4, 10], referred to as the wavefront resonance condition. This places a condition on the Rayleigh range that is of importance for FP-refractometry.

This constraint is based upon the expression for the radius of curvature of the wavefront of a Gaussian beam, given by Eq. (S3) above. Considering the fact that this curvature needs to be equal to the radii of curvature of the mirrors implies that, for a cavity comprising mirrors of which mirror i, placed at a distance z_i from the focus, which has a radius of curvature of R_i, (where i is 1 or 2) the wavefront resonance condition forces the Rayleigh range to be given by

$$z_R = \sqrt{z_i(R_i - z_i)}. \tag{S7}$$

This shows that the Rayleigh range of light locked to a mode of a cavity is solely and fully determined by the position of the mirrors and their curvatures. Hence, as long as these entities are not affected by the presence of gas, the Rayleigh range is also not affected by it.

The two most common types of cavities for FP-based refractometry are the symmetrical concave-concave and the plano-concave cavities.

A. A symmetrical concave-concave cavity

For a symmetrical cavity comprising two identical concave mirrors (i.e., a concave-concave cavity) with a radius of R separated by a distance L [11], the Rayleigh range can be written, in agreement with Kogelnik and Li [4], as

$$z_R = \frac{1}{2}\sqrt{L(2R - L)}. \tag{S8}$$

This implies that the single-pass Gouy phase for a laser beam addressing a longitudinal mode of the cavity, Θ_G, is given by

$$\begin{aligned}\Theta_G &= \arctan\left(\frac{L/2}{z_R}\right) - \arctan\left(\frac{-L/2}{z_R}\right) \\ &= 2\arctan\left(\frac{L}{\sqrt{L(2R-L)}}\right) \\ &= 2\arctan\left(\sqrt{\frac{L}{(2R-L)}}\right).\end{aligned} \tag{S9}$$

Utilizing a mathematical identity [12], it is possible to rewrite this as [13]

$$\Theta_G = \arccos\left(1 - \frac{L}{R}\right). \tag{S10}$$

This is in agreement with the expression for the single-pass Gouy phase used in the work by Silander et al. [14].

B. A plano-concave cavity

For a plano-concave cavity comprising a plane and a concave mirror, where the latter has a radius of curvature of R, separated by a distance L [15], and with the plane mirror placed in the focal plane of the beam, as utilized by Egan and Yang [16] as well as Yang et al. [17], the Rayleigh range can, in a similar manner, be written as

$$z_R = \sqrt{L(R-L)}. \tag{S11}$$

This implies that the single-pass Gouy phase, Θ_G, is given by [18]

$$\begin{aligned}\Theta_G &= \arctan\left(\frac{L}{z_R}\right) - \arctan(0) \\ &= \arctan\left(\frac{L}{\sqrt{L(R-L)}}\right) \\ &= \arctan\left(\sqrt{\frac{L}{R-L}}\right).\end{aligned} \tag{S12}$$

The last expression for the Gouy phase is in agreement with that stated in the work by Egan and Yang [16] as well as Yang et al. [17].

C. Conclusions

Since the Eqs. (S8) - (S12) are valid both in vacuum and in a medium with an index of refraction of n, as long as L and R are not affected by the presence of gas, i.e. for a non-deformable cavity, the Gouy phase is independent of the presence of gas.

However, as is shown in section 2.3 in the main text, for a deformable cavity, L and R are not independent of the presence of the gas. This implies that the Gouy phase acquires a pressure dependence as given by the Eqs. (7) and (8) in the main text.

3. DERIVATION OF AN EXPRESSION FOR ASSESSMENT OF REFRACTIVITY IN THE PRESENCE OF A PRESSURE-DEPENDENT GOUY PHASE

A. The phase resonance condition

As previously has been stated in the literature, the resonances of any optical cavity are determined by the condition that the round-trip phase delay of the light is a multiple of 2π. Based on this concept and the work by Koks and van Exter [19], Silander et al. [14] expressed a round-trip phase resonance condition for a FP-based system with DBR mirrors, addressing the m^{th} longitudinal mode of a FP cavity (but neglecting the influence of heating of the mirrors by the light), that reads

$$2k_{in}(L_0 + \delta L) + 2\phi - 2\Theta_G = 2\pi m, \tag{S13}$$

where k_{in} is the wave vector of the light in the cavity, L_0 the distance between the front facets of the two DBRs coatings of the mirrors in an empty cavity, δL the pressure-induced cavity deformation, ϕ the reflection phase of the DBR equipped mirrors (assumed to be identical), Θ_G the (single pass)

Gouy phase, and m the number of the longitudinal mode the laser addresses [20]. The subsequent derivation of an expression for the refractivity was then performed under the assumption that the Gouy phase was constant (i.e. independent of the presence of gas), leading to the Eq. (S10) in the Supplementary material of Ref. [14], which, in turn, was the basis for the Eqs. (13) - (16) in the same work. With the knowledge mediated in this work that the Gouy phase should have a component that is proportional to pressure, henceforth written as Θ_G^P, this derivation is redone below.

B. The pressure-dependent Gouy phase expressed in terms of index of refraction

For the derivation of an expression for the refractivity, it has been found convenient to rewrite the expression for the single-pass Gouy phase given by Eq. (8) in the main text in terms of a sum of a constant term and an index-of-refraction-dependent term, i.e. as

$$\begin{aligned}\Theta_G^P &= \Theta_G^0 + \Theta_G' P \\ &= \Theta_G^0 + \zeta\Theta_G'(n-1) \\ &= \Theta_G^* + \zeta\Theta_G' n,\end{aligned} \tag{S14}$$

where Θ_G^0 represents the pressure independent part of the Gouy phase, which for the two types of cavities addressed in this work is given by the Eqs. (S10) and (S12), where we have introduced ζ as a short-hand notation for $RT\frac{2}{3A_R}$, and where we, in the last step, have introduced Θ_G^* as a short-hand notation for $\Theta_G^0 - \zeta\Theta_G'$.

C. Under the condition that the laser is centered on the mirror center frequency and the heating of the mirrors by laser light is neglected

C.1. Derivation of an expression for assessment of refractivity in the presence of a pressure-dependent Gouy phase

A derivation of an expression for assessment of refractivity in the presence of a pressure-dependent Gouy phase can most conveniently follow that of the refractivity given in the work by Silander et al. [14]. This can most easily be performed by, in the expression for the frequency of the mode addressed by light in the presence of gas given by Eq. (S4) in the Supplementary material of that work, exchanging the Gouy phase, Θ_G, for its pressure-dependent counterpart, Θ_G^P.

This implies that the expression for the relative frequency difference between the modes addressed for an empty and a filled cavity [21], $\frac{\Delta\nu}{\nu_0}$, for the case when the laser is centered on the mirror center and, as was assumed above, and the heating of the mirrors by laser light is neglected, corresponding to Eq. (S7) in the Supplementary material of Ref. [14], can be written as

$$\frac{\Delta\nu}{\nu_0} = 1 - \frac{1}{n}\frac{m\left(1+\frac{\Theta_G^*}{\pi m}+\frac{n\gamma_c^*}{m}\right)}{m_0\left(1+\frac{\Theta_G^0}{\pi m_0}+\frac{\gamma_c}{m_0}\right)}\frac{1}{1+\delta L/L'}, \tag{S15}$$

where we have introduced γ_c^* as a short-hand notation for $\gamma_c + \frac{\zeta\Theta_G'}{\pi}$, and, as is stated in Ref. [14], where γ_c is defined as $2\tau_0\nu_c$ (where τ_0 is the group delay time for an empty cavity and ν_c is the center frequency of the working range), which, for an ideal DBR stack, is given by $\frac{1}{n_H - n_L}$, where n_H and n_L are the high and low indices of refraction of the DFB stack of the mirrors, respectively. L' is the effective length of the empty cavity comprising coated mirrors experienced during a scan, given by $L_0 + 2L_{\tau,c}$, where $L_{\tau,c}$ is the frequency penetration depth, given by $\frac{c\tau_0}{2}$. For details, see Silander et al. [14].

Following the procedure in the Supplementary material of Ref. [14], it is then possible to solve this expression for $n-1$. Doing so, yields first

$$n-1 = \frac{\frac{\Delta m}{m_0} + \frac{\Delta\nu}{\nu_0}\left(1+\frac{\Theta_G^*}{\pi m_0}+\frac{\gamma_c^*}{m_0}\right) - \frac{\delta L}{L'}(1-\frac{\Delta\nu}{\nu_0})\left(1+\frac{\Theta_G^0}{\pi m_0}+\frac{\gamma_c}{m_0}\right)}{(1-\frac{\Delta\nu}{\nu_0})\left(1+\frac{\Theta_G^0}{\pi m_0}+\frac{\gamma_c}{m_0}\right) - \frac{\gamma_c}{m_0} + \frac{\delta L}{L'}(1-\frac{\Delta\nu}{\nu_0})\left(1+\frac{\Theta_G^0}{\pi m_0}+\frac{\gamma_c}{m_0}\right)}, \tag{S16}$$

where Δm is the shift of the mode number, given by $m - m_0$.

Since δL, to first order, is proportional to pressure, and pressure, in turn, likewise, to first order, is linear with refractivity, it has been found convenient to introduce ε' as the refractivity-normalized relative elongation of the cavity due to the presence of the gas, defined as $\frac{1}{n-1}\frac{\delta L}{L'}$. As is shown in the Supplementary material to Silander et al. [14], ε' is an entity that has a very weak dependence on refractivity (for low pressures it acts as a constant and for higher it is weakly dependent on the refractivity) that can be written as $\varepsilon'_0\,[1+\xi_2(T)(n-1)]$, where ε'_0 is given by the low-refractivity value of ε', represented by $\kappa\zeta$, where κ is the deformation coefficient of the cavity, defined by $\frac{\delta L}{L'}=\kappa P$, hence being given by $\kappa RT\frac{2}{3A_R}$, and $\xi_2(T)$ is given by a combination of density and refractivity virial coefficients [22]. By doing this, and making use of the definitions of Θ^*_G and γ^*_c from above, it is then possible to rewrite Eq. (S16) as

$$\begin{aligned} n-1 &= \frac{\frac{\Delta\nu}{\nu_0}(1+\frac{\Theta^0_G}{\pi m_0}+\frac{\gamma_c}{m_0})+\frac{\Delta m}{m_0}}{1-\frac{\Delta\nu}{\nu_0}(1+\frac{\Theta^0_G}{\pi m_0}+\frac{\gamma_c}{m_0})+\frac{\Theta^0_G}{\pi m_0}+\varepsilon'_0-\varepsilon^{\delta R}_0+(n-1)\varepsilon'_0\,[1+\xi_2(T)]} \\ &\approx \frac{\overline{\Delta\nu'}+\overline{\Delta m'}}{1-\overline{\Delta\nu'}+\varepsilon'_0-\varepsilon^{\delta R}_0+(n-1)\varepsilon'_0\,[1+\xi_2(T)]}, \end{aligned} \tag{S17}$$

where, in the first step, the influence of the pressure-dependent Gouy phase has been expressed in terms of $\varepsilon^{\delta R}_0$, given by $\zeta\frac{\Theta'_G}{\pi m_0}$.

To simplify the expression, we have, in the second step, introduced $\overline{\Delta\nu'}$, given by $\frac{\Delta\nu}{\nu'_0}$ where, in turn, ν'_0 represents an "effective" empty cavity frequency, given by $\nu_0(1+\frac{\gamma_c}{m_0})^{-1}$, and $\overline{\Delta m'}$ is a short hand notation for $\frac{\Delta m}{m'_0}$ where, in turn, m'_0, is an "effective" mode number, is given by $m_0(1+\frac{\Theta^0_G}{\pi m_0})$. With the exception for the $\varepsilon^{\delta R}_0$-term, this expression is identical to Eq. (S.10) in Silander et al. [14].

It has been shown that when nitrogen or argon are addressed under the conditions pertinent to this instrumentation, the $(n-1)\varepsilon'_0\,[1+\xi_2(T)]$ term can, as long as pressures up to 100 kPa are addressed, safely be neglected [23]. In this case, Eq. (S17) can be written even more succinctly as

$$n-1=\frac{\overline{\Delta\nu'}+\overline{\Delta m'}}{1-\overline{\Delta\nu'}+\varepsilon'_0-\varepsilon^{\delta R}_0}. \tag{S18}$$

The Eqs. (S17) and (S18) both show, by the entities ε'_0 and $\varepsilon^{\delta R}_0$, that the pressure-dependence of the Gouy phase influences the refractivity in a manner similar to that of cavity distortion. The latter expression also shows that, when N_2 or Ar is addressed up to atmospheric pressures, since both $\varepsilon^{\delta R}_0$ and ε'_0 are index of refraction independent, $n-1$ can, by use of the ε-concept, be expressed in terms of a recursive-free expression.

D. For the case when the laser is not centered on the mirror center

In the case when the laser is not centered on the mirror center frequency, as has been shown by Silander et al. [14], it is possible to take this effect into account by simply exchanging γ_c for γ'_s, which is a short-hand notation for $\gamma_s\left(1+\frac{1+\chi_0}{1+\chi_1}\frac{\Delta\nu_{cs}}{\nu_s}\right)$ where γ_s is a short-hand notation for $\frac{2\tau_s(n)\nu_s}{n}$, which, in turn, under ideal conditions, is given by $\frac{1}{n_H-n_L}$, χ_0 and χ_1 are entities comprising coefficients to the non-linear response of the reflection phase with regard to the frequency deviation (from the mirror resonance frequency) while $\Delta\nu_{cs}$ represents the difference in natural frequency between the mirror center frequency and the center frequency of the working range, defined as $\nu_c-\nu_s$. In this case, the same expressions for refractivity as above is valid, i.e. the Eqs. (S17) and (S18), with the alteration that ν'_0 is given by $\nu_0(1+\frac{\gamma'_s}{m_0})^{-1}$. See Silander et al. [14] for details.

E. In the presence of heating of the mirrors by laser light

As was recently mediated by Zakrisson et al. [24], when pressures are assessed in the viscous pressure region, absorption of laser light in the mirrors will give rise to a small alteration in the proportional response and a pressure-independent offset, where the latter is significant for He but considerably smaller for Ar and N_2. This implies that, in the presence of heating of the mirrors by laser light, Eqs. (S17) and (S18) need to be complimented by the factor $\left(1-\frac{a_{mP}}{b+P}\right)$. See Zakrisson et al. [24] for details.

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9. Or, alternatively its intensity (or irrradiance) at the origin, I_0, which is given by $\frac{\varepsilon_0 c}{2}|E_0|^2$, where ε_0 and c are the permittivity of free space and the speed of light, respectively.
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12. $\arctan(x) = \frac{1}{2}\arccos\left(\frac{1-x^2}{1+x^2}\right)$.
13. Utilizing the mathematical identities $\arccos(x) = \arcsin\left(\sqrt{1-x^2}\right) = \arctan\left(\sqrt{\frac{1-x^2}{x^2}}\right)$, this can alternatively be written as $\arcsin\left(\sqrt{\frac{L(2R-L)}{R^2}}\right)$ or $\arctan\left(\sqrt{\frac{L(2R-L)}{(R-L)^2}}\right)$, respectively.
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