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Comprehensive three-dimensional ray tracing model for three-mirror cavity-enhanced spectroscopy

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A 3D ray tracing model is used to simulate optical reinjection in a nonresonant optical cavity, for off-axis integrated cavity output spectroscopy. The optical cavities are optimized for maximum intensity enhancement factors via a grid search and a genetic algorithm. Intensity enhancement factors up to 1400 are found for short cavities (3 cm) and up to 101 for long cavities (50 cm). The model predicts that short absorption cells can be used, having a long effective path length and a high throughput power. This opens new opportunities in the field of ultrasensitive absorption spectroscopy and allows the design of compact optical gas sensors. © 2018 Optical Society of America

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1. INTRODUCTION

Cavity-enhanced absorption spectroscopy (CEAS) allows sensitive absorption spectroscopy to quantify weak molecular transitions or to detect trace gases, down to the parts-per-billion volume (ppbv) mixing range. In combination with high-resolution spectroscopy, it also enables high specificity in complex gas mixtures [1–4]. Cavities utilizing high-finesse optical resonators provide effective interaction path lengths, up to a path length of several kilometers. In 1988, cavity ring-down spectroscopy (CRDS) was the first cavity-enhanced method that demonstrated high sensitivity for molecular absorption spectroscopy, using the decay time of the cavity [5]. Once in resonance with a cavity mode, the light enters the optical cavity and builds up a standing wave between the two mirrors. The light is trapped and after switching the laser off, the intensity rings down by leaking out through the front and back mirror of the cavity. In 1998, integrated cavity output spectroscopy (ICOS) was proposed as a simple, sensitive and robust approach to measure direct absorption of gases, rather than via a decay time [6–9]. When the light beam is aligned on-axis in the cavity, the output signal highly depends on the efficient in-coupling of the laser frequency into the cavity mode; its throughput is fluctuating accordingly. For a high-finesse cavity

the linewidth of the cavity transmission peak is in the order of kilohertz; laser linewidths do not reach this number easily. As such, a feedback loop for laser stabilization is needed to lock the laser to the cavity to get near to 100% transmission. Attempts to average the cavity transmission without feedback by modulating its length generally lead to amplitude noise [10,11]. Another approach for efficient injection of the light into optical cavities is using optical feedback in ν -shaped cavities [12]. In such a configuration, the laser is locked to the cavity by optical feedback from the cavity into the laser. The laser wavelength and one cavity mode get synchronized, leading to efficient coupling.

Alternatively, the laser light can be injected in an off-axis (OA) configuration, thereby reducing amplitude noise at the cost of intensity transmission through the cavity [8,11,13–16]. In this situation the laser spot in the cavity is not coming back directly to its entry spot after one round trip ($s = 2L$, with s being the laser path length and L the cavity length), but after a number of round trips ($s = 2mL$, with m the number of round trips). As a result, the free spectral range (FSR) of the cavity is decreased by a factor m , leading to an m -time higher density of cavity modes. When the laser line width is broader than this FSR, the laser will now couple to multiple cavity

modes at once. This causes the transmission spectrum of the cavity to become virtually flat. The coupling of laser light into the optical cavity no longer depends on the wavelength of the incoming beam.

Such an OA alignment leads to lower intensity noise levels and therefore an increased signal-to-noise ratio. In addition, the OA alignment is less sensitive to small changes in the input angle or position of the laser beam, making the total system more robust and easy to implement even under extreme conditions. A comprehensive overview of how to enhance the sensitivity in OA-ICOS can be found elsewhere [7,8,11].

A number of factors affect the performance of OA-ICOS. In general, mid-infrared detectors have typically 1 to 3 orders of magnitude lower detectivity (D^*) compared to near infrared detectors (see, e.g., [17]). As such, higher laser intensities are needed onto the detector in order not to be limited by the detector noise. With a 99.98% reflectivity of the mirrors from the absorption cavity, the transmitted intensity will be $\sim 0.01\%$ of the total incoming intensity; the rest will be reflected backward and is lost [8,18,19]. The sensitivity enhancement for weak absorptions is highly dependent on the effective path length of such cavities, $L_{\text{eff}} \approx 2L/(1-r)$, with L the cavity length and r the reflectivity of the mirrors. The factor 2 accounts for laser radiation leaking out on both sides of the high-finesse cavity. As a result, owing to the lower detectivity of mid-infrared detectors, it is difficult to get high absorption sensitivity using high-finesse optical cavities, with high-reflective mirrors.

A way to couple more light into an optical absorption cavity is to reinject the back-reflected light from the first cavity mirror with a third mirror in front of the first cavity mirror. Initially, the laser light enters through a hole in the reinjection mirror. O'Keefe and coworkers reported such optical reinjection by showing a transmission intensity enhancement of 22.5. A ray tracing model predicted that an enhancement factor of 25 can be achieved [20]. At the same time, Centeno *et al.* measured an enhancement factor of 28, leading to a tenfold increase in the signal-to-noise ratio [18,21]. They also introduced an optical ray tracing model, using matrix-based paraxial approximations. This study was restricted to a limited number of parameters, such as the mirror radii of curvature (R), cavity length (L), reinjection cavity length (d), position and size of the entrance hole at the reinjection mirror, and incoming angles of the laser beam [18]. It was found that the number of round trips before coming back to its original position inside an optical cavity can be increased significantly by taking into account astigmatism.

Methods for tracing rays through an optical system containing mirrors and lenses are well recognized. For example, geometrical optics in combination with matrix-based methods was used to model Herriott cells, multiple-reflection optical cells, and laser resonator designs [22–26]. Nowadays, optimal designs can be calculated with commercial software. However, one of the main drawbacks of such software is the lack of a proper starting configuration; this leads to unsatisfactory optimization of the nonlinear optical system [27].

In this work, we used a 3D ray tracing model to simulate ray traces in our system. With the model we aimed to maximize the

intensity through the cavity and the absorption path length. For this, we used two mathematical approaches: a grid search and a genetic algorithm (GA). The grid search calculated the influence of the reinjection mirror, by changing a number of parameters in discrete steps: the position of the entrance hole, angle of the incoming laser beam, reinjection and absorption cavity length, and mirror radii of curvatures. In contrast to the grid search, the GA varies the parameters as a continuous (nondiscrete) function toward a global optimal solution for the entire search space [28]. As the grid search demands long computational times, the GA is much more time efficient for calculating the optimal configuration.

The goal of this work is to find a maximum intensity enhancement by inserting a third mirror. For this, we defined an enhancement factor: the intensity ratio of transmission with a reinjection mirror to a situation without a reinjection mirror. We investigated the configurations and the tolerances around the optimal configurations, using the gain factor as a criterion for an optimal configuration.

2. 3D RAY TRACING MODEL

Ray tracing is the propagation of the light rays through successive optical interfaces of an optical system. It can be performed in various steps. The first step involves the travel of the ray over a distance inside a medium toward a point of intersection at an optical surface; the second step is the calculation of the surface normal at this intersection point; and the third step is the transmission/reflection interaction at this surface. For the ray tracing calculations, the selection of a proper mathematical model plays a key role. By using a 3D ray tracing model, we wanted to incorporate the effects of large entrance angles and strongly curved spherical surfaces that, among others, lead to astigmatism [29], thereby going beyond the classical paraxial theory.

Here, we use a model that provides exact calculations in 3D, a contrast to classical matrix-based calculations for ray-surface interactions from object point to image point [30]. For a valid ray tracing model, complete information of the optical system under study is needed: surface type and shape (plane, spherical, parabolic, etc.), radius of curvature, size of the boundary of the surface, position and orientation of the surfaces, and refractive indices of the materials. As algorithms for performing 3D skew ray tracing in optical systems are well established, we developed and implemented these algorithms from earlier work, e.g., by Spencer and Murty [31], and Lindlein [32].

A general illustration of the beam propagation through the optical system is shown in Fig. 1. Starting at the entrance hole in mirror M_1 (reinjection mirror), the optical beam travels over a distance d to mirror M_2 . Mirrors M_1 (concave) and M_2 (convex) form the reinjection cavity, while mirrors M_2 (now concave) and M_3 (concave) form the absorption cavity with length L . A small entrance hole (typically 1 mm diameter) in mirror M_1 provides an entrance for the incoming laser beam.

In this situation the thickness of mirror M_2 is not taken into account, just like the surface coatings of the mirrors. In more detail, mirror M_2 has an antireflective (AR) coating at the reinjection cavity side and a high reflective (HR) coating on the

Fig. 1. Ray tracing through the optical system formed by the re-injection and absorption cavity with length d and L , respectively. The three spherical mirrors ($M_{1,2,3}$) have radii of curvatures ($R_{1,2,3}$). The red spots show the round trips in the reinjection cavity, the blue spots in the absorption cavities. The resulting light is imaged via a lens onto the detector. The entrance angles α_x and α_y are shown for the incoming laser beam. The entrance hole is at a distance $\sqrt{x^2 + y^2}$ from the central axis.

absorption cavity side. At the HR coating side, the ray is split in a reflected ray and a transmitted ray; their intensities depend on the coating reflectivity (e.g., 99.98%). The reflected ray from M_2 will make n round trips in the reinjection cavity. The initial entrance angles α_x and α_y through the hole at mirror M_1 are defined as the angles between the ray and the space coordinates in x and y directions. The entrance angles into the hole and the x, y position of the hole are critical. It should be avoided that the incoming laser exits back through the entrance hole already after a few round trips in the reinjection cavity. The transmitted ray at mirror M_2 covers a distance L , before it interacts with mirror M_3 , after which it makes m round trips in the absorption cavity in an OA configuration.

The 3D ray tracing model is implemented in Matlab, and a number of steps are followed in a continuous flow process, as shown in Fig. 2. Panel (a) shows the ray tracing interacting with the three mirrors. The initial ray enters the reinjection cavity via the hole in M_1 to M_2 . At M_2 the ray is entering the mirror via the AR coating on that side of the mirror, and the ray is refracted in the mirror material and travels toward the next optical surface of M_2 . At this surface the ray is reflected/transmitted, taking into account the HR coating at this mirror surface. The reflected ray travels back to the AR side of the mirror, is refracted, and travels to M_1 .

Within Matlab, the properties of the spots at the HR side of M_2 (size, intensity, position, and angle) resulting from the ray traveling through the reinjection cavity are stored. After this, in a separate procedure, the ray traveling in the absorption cavity is calculated. From each of the previous determined spots on M_2 the ray travels in the absorption cavity to M_3 where it is reflected, traveling back to M_2 , etc.

The procedure to calculate the ray properties towards the next surface is presented in Fig. 2(b). At the start, the ray has a position, angle, and intensity. In step (I), the intersection of the ray with the surface is determined. The program continues to trace the ray if it is on the active part of the mirror; otherwise, it stops. In step (II) the actual ray-surface

Fig. 2. (a) Diagram of the ray tracing through the four surfaces of the three mirrors. Initially, the ray tracing in the reinjection cavity is calculated and properties on the HR surface of M_2 are stored. After that, from each spot from that surface the rays are traced in the absorption cavity. Panel (b) is a diagram of the interaction of each ray with a mirror surface and its travel to the next optical surface calculating its properties (size, intensity, position, and angle).

intersection is calculated, while in step (III) the normal to the surface is evaluated. The properties of the reflected/refracted beam are calculated and saved, as well as the beam spot positions and intensity [step (IV)]. If the beam intensity is larger than a predefined threshold value, the procedure continues.

The threshold value for the laser intensity is set to 10^{-4} of the initial laser intensity. After each reflection in the reinjection cavity, a small portion of the laser intensity is lost either to the absorption cavity or backward through M_1 . The subroutine is ended when the laser intensity is lower than the threshold value. Since we model the spot size with one central ray and six rays (see discussion below) on the circumference of the laser spot, the calculation is also stopped if one of the rays misses the mirror surface. Since M_1 is a HR mirror, the back transmitted laser intensity through the reinjection mirror M_1 is not considered in the calculations. For a mirror reflectivity of 99.98%, the output intensity on the detector will be reduced by a factor 10^{-4} . To define the enhancement factor, we calculated the intensity with and without the reinjection mirror

on mirror M_3 . Adopted from [8], the enhancement factor can be written as (assuming no absorption)

$$P_{\text{enhanced}} = \frac{I^{3\text{-mirror}}}{I^{2\text{-mirror}}} = \frac{\sum_{i=0}^n r^{2i} m_i}{m_0}, \quad (1)$$

in which $I^{3\text{-mirror}}$ and $I^{2\text{-mirror}}$ are the intensities with and without the reinjection mirror; n is the number of round trips in the reinjection cavity; m is the number of round trips in the absorption cavity; r is mirror reflectivity; m_i is the number of round trips in the absorption cavity from spot i on the HR surface of M_2 ; and m_0 is the transmission with $i = 0$ (without the reinjection mirror).

3. GRID SEARCH AND GENETIC ALGORITHM

To perform a grid search with the 3D ray tracing model, seven variables are encoded within the script: the entrance hole coordinates x, y (0 mm to 30 mm, with step size of 1 mm); angles of the incoming laser beam to the surface normal α_x, α_y (-6° to 6° , step size 0.1°); reinjection cavity length d ; absorption cavity length L ; and radii of curvature of the mirrors ($R_{1,2,3} = R$). The radii are set equal, as one variable. Four parameters are kept constant throughout the simulations, namely the reflectivity of the mirrors (99.98%), threshold intensity value (10^{-4}), diameter of the entrance hole (1 mm), and mirror diameter (2 inch = 50.4 mm).

Using the grid search approach any configuration can be simulated by varying systematically the parameters $R_{1,2,3}, d$, and L in discrete steps and optimizing the entrance hole and angles. However, these simulations are time-consuming as there are many variables. For example, keeping $R_{1,2,3}$ at a fixed value, such an optimization took several days, using Matlab, version R2015a, 64-bit, installed on a PC with an Intel(R) Xeon(R) CPU (32 units, 3.20GHz) with 264 GB RAM. To reduce these time-consuming calculations, we investigated the applicability of GA for efficiently finding optimal configuration parameters. Indeed, the GA significantly reduced the optimization time: for a fixed $R_{1,2,3}$, the calculation, to find a maximum enhancement factor, took 1.2 h repeating this 10 times.

The GA generates an initial population of parameters and evaluates them using a fitness value. In our case, the fitness value is the maximum intensity transmitting to the detector. The parameter combinations with the highest fitness values are selected as “parents” to form a new generation; parameter combinations with low fitness values are discarded. The parents with the highest fitness value are “elites” and passing on unchanged to the next generation. For other members of the next generation crossover is used to generate new members from the parent population, with a crossover value of 0.5. Third, new members are created by mutation, in which small, random changes are made to each of the values. All these new members form the new generation, and each member is evaluated using the fitness function. This process repeats until a stop condition is reached. This stop condition is either a maximum number of generations (200 to 600 generations) or a number of generations during which no change occurs. Since the generations are randomly generated with restricted conditions for crossover and mutation, the GA does not always generate the same output.

With using the GA, the set of parameters were slightly changed. Owing to cylindrical symmetry of the starting conditions of the incoming laser beam, y is set to 0 mm and only positive values of α_y are considered. Compared to the grid search local optima are found very efficiently. The calculation stops once the intensity drops below the threshold value, goes back through the entrance hole, or crosses the mirror edge. For the GA calculations, the radii of curvature of the reinjection mirror R_1 were allowed to be different from those of the absorption cavity mirrors ($R_{2,3} = R$).

4. RESULTS AND DISCUSSION

Initially, we investigated with the grid search, the symmetrical character of the setup for the radii of the curvatures of the three mirrors: $R_{1,2,3} = R$. Each mirror $R_{1,2,3}$ was varied from 0.1 m to 10^3 m; the latter value is used to simulate a flat mirror. During these preliminary investigations, it was observed that symmetrical optical configurations ($R_{1,2,3} = R$) give consistent higher enhancement factors (data not shown); this was in agreement with literature data [33,34]. Because of this, mainly configurations with equal radii of curvatures have been considered with the grid search approach.

A. Spot Pattern in Reinjection and Absorption Cavity

Long effective path lengths are the key interest in OA-ICOS, CEAS, and CRDS, or any other cavity-based spectroscopy method. This can be achieved by increasing the number of round trips in the absorption cavity and maximizing the transmission intensity through the absorption cavity.

For this, in our case we need a maximal number of spots on the mirrors. An example of calculated spot patterns is shown in Fig. 3 using the parameters for a short length of both cavities: $d = 7$ cm, $L = 5$ cm, $R_{1,2,3} = 10$ cm, $r = 99.98\%$, $\alpha_x = 0.3^\circ$, $\alpha_y = 0.7^\circ$, $x = 10$ mm, and $y = 0$ mm. In Figs. 3(a) and 3(b), an elliptical shape is formed (with 93 spots) on both mirrors (M_1 , concave and M_2 , convex) of the reinjection cavity. Compared to M_1 , the elliptical shape is reduced in

Fig. 3. Example of spot patterns on mirrors M_1 and M_2 in the reinjection cavity (panels a, b) and absorption cavity (panels e, f) for a specific parameter set (for parameters, see text). The spot pattern in the absorption cavity is created by the first ray (with the highest intensity) entering from the reinjection cavity. Each further ray (spot) from the reinjection cavity is creating a similar spot pattern in the absorption cavity (not shown). Panels (c, d), and (g, f): calculated spot patterns in the reinjection cavity and absorption cavity using an optimal configuration generated by the GA (for parameters, see text).

size on mirror M_2 , because the incoming ray through the entrance hole is entering at an angle. The spot pattern on the mirrors M_2 and M_3 of the absorption cavity (panel e, f) are resulting from the first ray entering from the reinjection cavity into the absorption cavity. It can be seen that there are 7489 spots formed in the absorption cavity from this first ray. Each further ray (spot) from the reinjection cavity is creating a similar spot pattern in the absorption cavity (not shown). Owing to the threshold condition, the number of spots for these rays in the absorption cavity is slightly decreased, as the spot intensity from the reinjection cavity is decreased. Using the GA, a better configuration was found with slightly changed initial parameters ($d = 7.013$ cm) but stronger entrance angles ($\alpha_x = -5.8^\circ$, $\alpha_y = 5.7^\circ$). Large entrance angles result in an astigmatic effect, resulting in a spot pattern of a Lissajous figure [35,36]. Panels (c, d), and (g, f) show the calculated spot patterns in the reinjection cavity and absorption cavity, respectively. The GA found 864 spots in the reinjection cavity compared to 93 spots for the standard condition. Within the calculations, we do not take into account the back transmission from the absorption cavity into the reinjection cavity due to the high reflectivity of mirror M_2 ($r = 99.98\%$). The number of spots in the absorption cavity is the same for both configurations (7489 spots) due to the threshold condition.

Shape and location of the spots strongly depend upon the radii of curvature of the mirrors and the distance between the mirrors, d and L . In the reinjection cavity, the optical ray fulfills the condition for exiting via the entrance hole when [8,14,23]

$$m2(\Theta) = 2\pi n, \quad (2)$$

with m the number of round trips, n an integer, and Θ the rotation angle per pass given by

$$\cos(\Theta) = 1 - (d/R). \quad (3)$$

The spot patterns look similar to drawings produced by a spirograph and are termed hypotrochoids; see, e.g., [37]. The spot patterns were well explained by Herriott and coworkers in 1964 [35]. Krzempek *et al.* [38,39] reported dense patterns in their multipass cell after minimizing the etalon fringe effects. Optimized spot patterns cover most of the mirror's surface with an empty center in the middle of the mirror, as in our case. Ellipses of equal magnitude in each direction tell about the least effect of astigmatism, providing the advantage of being very tolerant in optical alignment.

Initially, our calculations used a single ray to represent a single laser beam through the optical system. However, the dimensions and shape of the beam cannot be ignored; they change upon each reflection by the mirrors. As such, it does not necessarily mean that the full beam will be reflected, as a part of the beam can cross over the edge of the mirror. To account for this, the built-in function is modified by simulating the circumference of the laser beam with six points around the center point, yielding a total calculation of seven rays for the shape of the beam. For the initial beam, the six spots are on a circle (diameter 1 mm). Upon multiple reflections, the beam shape is distorted over time, as is illustrated with an example in Fig. 4 (spots on reinjection mirror M_1).

The maximum number of reflections is reached when the beam exits the reinjection cavity through the entrance hole

Fig. 4. Typical spot-pattern on the reinjection mirror M_1 , after multiple reflections. Here, the laser beam is represented by seven spots. The entrance hole is represented by a small circle near the center. 1st spot (red), 21st spot (dark blue), 41st spot (green), 61st spot (brown), 81st spot (magenta), and 101st spot (cyan). Only five spots of the 101st spot (cyan) are visible, indicating that part of the beam is crossed at the edge of the mirror, meaning that the maximum number of reflections is obtained.

or when it crosses the edge of the mirror. For the latter situation the algorithm stops. When the ray exits via the entrance hole, it is difficult for the GA to find a better solution. When the beam leaves the cavity through the entrance hole, a small change of the ray position within the entrance hole does not change the fitness value. To better parameterize this situation, we have incorporated the position of the ray in the exit hole into the optimization. From the spot position in the entrance hole we made a continuous function, represented by

$$n' = n + \frac{k}{b}, \quad (4)$$

where n' represents a modified spot count, which is used to calculate the enhancement factor, n is the spot count (an integer), k is the distance between the position of the ray and the center of the entrance hole, and b is the radius of the entrance hole. Because k/b is always between 0 and 1, the fitness value will now increase gradually as the solution gets closer to increasing the spot count (i.e., closer to the edge of the hole).

B. Enhancement Factor Using the Grid Search

Using the grid search approach, an enhancement factor for the intensity was calculated for nine radii of curvature of the mirrors (between $R = 10$ and 100 cm), as shown in Fig. 5. For each R , the reinjection cavity length d is varied, such that d/R is between 0.1 and 1 (step size 0.1). For the absorption cavity, three cavity lengths are considered: a more planar configuration with $L = R/2$ (red triangles); a confocal configuration $L = R$ (black circles); and $L = 30$ cm (blue squares).

For a fixed absorption cavity length $L = 30$ cm and at small R (Fig. 5 panels (a, b), blue squares), almost no enhancement is observed, irrespective of the d/R ratio. In this case, we have an unstable absorption cavity. This comes directly from the stability criterion of optical cavities [40], given by

Fig. 5. Calculated enhancement factor as a function of the d/R ratio using the grid search approach. Panels (a–i) show increasing radius of curvature $R_{1,2,3} = R$ (from 10 to 100 cm). Each panel contains three situations: red triangles ($L = R/2$, nearly planar), black circles ($L = R$, confocal), and blue squares ($L = 30$ cm).

$$0 \leq \left(1 - \frac{L}{R_1}\right) \left(1 - \frac{L}{R_2}\right) \leq 1. \quad (5)$$

For $L = 30$ cm and with an increasing radius of curvature, the enhancement factor increases if it comes closer to a near planar situation [Fig. 5 panels (d–i)]. At this point, a maximum value is found at $d/R = 0.7$ (enhancement factor ~ 70).

It is remarkable that for all situations ($L = R/2, L = R, L = 30$ cm) a minimum enhancement factor is reached at $d/R = 0.5$. Recalculations with simple ray tracing matrices (with no astigmatism) showed that under these conditions the ray exits the reinjection cavity already after six round trips through the entrance hole, irrespective of its entrance angle and position.

In Fig. 5 panel (c), $L = R = 30$ cm gives identical enhancement factors for two approaches; the same goes for Fig. 5 panel (e) the near planar situation ($L = R/2 = 30$ cm). In the case of the confocal situation ($L = R$), the absorption cavity is at the edge of stability, possibly explaining the quite fluctuating enhancement factor results. For the near planar situation, $L = R/2$, a more consistent pattern can be found in all panels of Fig. 5, with a maximum enhancement factor at $d/R = 0.7$, having optimal entrance angles (α_x, α_y) and entrance hole coordinates (x, y). For the latter situation ($L = R/2, d/R = 0.7$), Fig. 6 shows the enhancement factors for $R = 30$ and 100 cm as a function of entrance angles α_x, α_y , using an optimized position of the entrance hole.

At longer cavity lengths, maximum enhancement factors are in between very narrow boundaries of the entrance angles, Fig. 6 panel (b).

Figure 7 shows, with cylindrical symmetry, the enhancement factors depending on the position of the entrance hole using the same configurations as in Fig. 6. The color code represents the enhancement factor and shows that for $R = 30$ cm

Fig. 6. Enhancement factor as function of entrance angles (α_x, α_y) for the beam passing through a 1-mm entrance hole at an optimized position ($L = R/2, d/R = 0.7$). Panel (a) $x = 10$ mm, $R = 30$ cm; panel (b) $x = 18.5$ mm, $R = 100$ cm.

Fig. 7. x, y coordinates of the 1-mm entrance hole with the resulting enhancement factor for the same configurations as Fig. 6: panel (a) $R = 30$; panel (b) $R = 100$ cm. Enhancement factors are shown in color code.

easily a maximum enhancement factor can be found within a radius of 15 mm from the center. At larger R [panel (b)], fluctuation of the enhancement factor at the edge of the ring can be observed. This effect is a consequence of the discrete nature of the grid search. The fluctuations are caused by the limited step size of the spatial coordinate (0.1 mm) and input beam angle (1°). This also indicates that it is comparatively more difficult (the initial parameters need more precision) to get experimentally a maximum enhancement factor using larger cavity lengths and larger radii of the curvature.

A brief summary of the simulations for different optical cavity parameters are given in Table 1 for $L = R, L = 30$ cm and $L = R/2$, varying R between 10 and 100 cm with $d/R = 0.7$; the latter is the value at which (mostly) a maximum enhancement factor is reached (see Fig. 5). For given values of R, d , and L : the x -coordinate of the hole and the entrance angles α_x, α_y are calculated together with the maximum enhancement factor and the number of spots on the reinjection mirror.

A large enhancement factor gives a high throughput in laser intensity. However, this can be reached using very short cavities, resulting in short absorption path lengths and reduced

Table 1. Brief Summary of the Simulations Using the Grid Search, for the Optical Cavities Parameters Configurations of $L = R/2$, $L = 30$ cm, and $L = R$, Varying R between 10 and 100 cm with $d/R = 0.7^a$

	R (cm)	$d(\Delta)$ (cm)	$L(\Delta)$ (cm)	$x(\Delta)$ (mm)	$\alpha_x(\Delta)$ (deg)	$\alpha_y(\Delta)$ (deg)	Enhancement Spots Factor	Scaled Enhance. Factor (m)
$L = R/2$ Near planer absorption cavity	10	7.0 (0.2)	5(4)	10 (1)	0.3 (0.2)	0.7(0.3)	93	4.4
	30	21.0 (0.3)	15 (15)	10 (2)	-1.7 (0.35)	2(< 0.01)	77	10.8
	50	35.0 (0.2)	25 (25)	10 (2)	-1.1 (0.15)	1.2(0.01)	77	18
	70	49.0 (0.3)	35 (34)	10 (0.2)	-0.5 (0.01)	0.8(< 0.01)	77	25.2
	90	63.0 (0.05)	45 (39)	10.0 (0.1)	-1.1 (0.01)	0.5(< 0.01)	62	34.6
	100	69.85(0.3)	50 (41)	17.8 (3)	-1.1 (0.01)	0.5(0.01)	64	38.5
$L = 30$ cm Absorption cavity length	10	7(< 0.01)	30 (28)	10 (0.5)	0 (0.4)	0 (2)	1	0.10
	30	21 (0.3)	30 (22)	10 (2)	-1.7 (0.4)	2.0(< 0.01)	21	5.7
	50	35 (0.2)	30 (24)	10(4)	-1.1 (0.15)	1.2 (0.01)	77	21.6
	70	51.3 (0.1)	30 (48)	10 (2)	-1.3 (0.01)	-0.1(0.6)	77	21.6
	90	63 (0.4)	30 (43)	10 (0.05)	-1.1(< 0.01)	0.5(< 0.01)	47	14.1
	100	70 (0.4)	30 (50)	10 (0.7)	-0.5 (0.04)	0.6(< 0.01)	64	21.6
$R = L$ Confocal absorption cavity	10	7 (0.2)	10 (8)	10 (2)	-5.1 (0.5)	-5.0 (0.5)	12	0.4
	30	21 (1)	30 (22)	10 (2)	-1.9 (0.4)	-2.0 (0.04)	22	5.7
	50	35 (0.1)	50 (35)	10 (2)	-1.3 (0.2)	1.2 (0.01)	32	14
	70	49 (0.3)	70 (50)	10(2)	-0.5(< 0.01)	0.8(< 0.01)	32	22.4
	90	63 (0.5)	90 (48)	10 (0.05)	-1.1(< 0.01)	0.5(< 0.01)	22	18
	100	70 (0.5)	100 (67)	10 (0.7)	-0.5 (0.04)	0.6 (0.01)	38	36

^aOptimal parameters found are x , α_x , and α_y , with their spot number and enhancement factor. The scaled enhancement factor is the product of the enhancement factor and L (in meters). The Δ value in brackets is the width of the parameter before the enhancement factor drops to half of its value.

sensitivities. To compare short cavities with large enhancement factors and long cavities with low enhancement factors, we introduced the scaled enhancement factor. The scaled enhancement factor is a product of the enhancement factor and the absorption cavity length L (in meters). As such, it is a compromise between detector sensitivity and detection sensitivity.

The value in between brackets (Δ) after a parameter is the full width over which the enhancement factor stays above 50% of its maximum value. As such, it represents the experimental flexibility in alignment within the cavity before losing enhancement. From this, it can be seen that the length of the absorption cavity L is quite insensitive for the enhancement factor (as expected), and d is quite critical within a few millimeters. The x coordinate of the entrance hole is in all cases almost the same value, except for $R = 90$ and 100 cm, in which case it is close to the edge of the reinjection mirror. For short radii of curvature, the acceptance angle for α_x and α_y is wider than for long cavities with large radii of curvature.

The grid search gave a global view of the fitness landscape: a more fine-grained view was not achievable, due to the long calculation times needed.

C. Enhancement Factor Using GA

Using the GA, the parameters are varied as continuous functions, to achieve optimal solutions in the entire search space. Initially, the enhancement factor was used as fitness value (later the scaled enhancement factor was used as fitness value, see below). The algorithm had as free parameters the following: the entrance hole coordinate x (between 0 and 30 mm); entrance angles α_x , α_y (between -6° and 6° , and 0 and 6° , respectively); the reinjection cavity length d ; absorption cavity length L ; and radii of curvature of the mirrors, here $R_1 \neq R_2 = R_3$. The calculations are performed with 2-inch diameter mirrors having reflectivity of 99.98%.

As the GA can be stuck in local minima, we tested different combinations of population size, crossover, and mutation values to obtain optimal solutions and a balance between exploitation and exploration in an acceptable time. Owing to the random nature of the GA, we repeated the GA for each situation 10 and 20 times to investigate the reproducibility. The results were always verified, performing a narrow fine-grained grid search around the GA optimum.

Using the enhancement factor as fitness function, the results of applying ten times the GA are shown in Figs. 8(a)–8(f). For every enhancement factor (panel a) the other parameters values found are shown: x (panel c); α_x , α_y (panel d); d and L (panel e); and R_1 and $R_2 = R_3$ (panel f). In this situation the scaled enhancement (panel b) is directly calculated from the product of the enhancement factor and the absorption cavity length L , as described in Section 4.B.

For larger x values (panel c) a larger enhancement factor is achieved, due to more OA configurations of the rays; a phenomenon well understood. R_1 [reinjection mirror radius, panel (f)] shows a stronger radius of curvature than the absorption cavity mirrors ($R_{2,3}$); as such, it is a stable cavity [see Eq. (5)]. The results show that, using the enhancement factor as fitness value, none of the parameters are consistently reproduced, although very high values can be obtained. The reason for this is that the optimization landscape is very discrete, resulting in situations in which the GA can be trapped in local optima.

In some of the runs, a high enhancement factor is obtained for short absorption cavities (a few centimeters long). Despite the high enhancement factor, such a configuration cannot be in favor of high absorption sensitivity. With a 30-cm-long absorption cavity and mirror reflectivity of 99.98%, the effective path length is 3 km.

With a 3-cm-long cavity, the effective path length will be 300 m, resulting in a 10 times less strong absorption.

Fig. 8. Results of applying 10 times the GA, using the enhancement factor as fitness function, as shown in panel (a). For each optimum the other parameters are: x (panel c), α_x, α_y (panel d), d and L (panel e), and R_1 and $R_2 = R_3$ (panel f), with the calculated scaled enhancement in panel (d). Panels (f–l) give the results if the scaled enhancement factor is used as the fitness function in the GA (20 repetitions).

Ideally, both the total intensity and the effective path length should be as high as possible.

Therefore, we optimized the approach by using the scaled enhancement factor as fitness function (product of enhancement factor and absorption cavity length L). The results from this new approach are presented in Fig. 8 (panels g–l). The calculations for the scaled enhancement factor were repeated 20 times for reproducibility. At this point, both cavity lengths (L and d) became reproducible having lengths of about 50 cm (panel k).

Using the enhancement factor as fitness function, strongly varying enhancement factors were found (panel a) with values up to 1400 corresponding to a calculated scaled enhancement factor of 44 (panel b). Applying the scaled enhancement factor as fitness function, the results became reproducible, with values

for the scaled enhancement factor from 38 to 58 (enhancement factors 75 to 110). This illustrates that despite the far lower enhancement factors the longer absorption cavities can be more efficient in absorption strength.

Using the scaled enhancement factor as fitness function, there is an upper limit to it. This can be explained as follows: without astigmatism, the spots on the reinjection mirror lie on an ellipse; i.e., periodically, the spots end up in the same location as before. With astigmatism, the ellipse rotates slightly, and this discrepancy increases as the mirrors become more astigmatic. Therefore, with mirrors that are less curved the light will eventually end up in the entrance hole of the reinjection mirror. For long cavities, the astigmatism is not strong, and the spot pattern will be mostly along a single ellipse, resulting in an upper limit to the number of spots on the reinjection mirror and, thus, an upper limit to the enhancement factor.

To reduce the calculation time for the optimal configuration of the parameters, we investigated whether we could omit the calculation of the spot patterns in the absorption cavity. For this, we correlated the number of spots on the reinjection mirror with the enhancement factor. We assumed that all rays are reflected an infinite number of times in the absorption cavity with a 100% transmission of intensity through the absorption cavity. Figure 9 shows a relationship between number of spots on the reinjection mirror and the enhancement factor. For this comparison, we used a reinjection mirror diameter of 50.8 mm and a mirror diameter of 25.4 mm for the absorption cavity; similar figures were obtained when all the mirror diameters were equal. From Fig. 9 it can be seen that there is a proportional relationship, although deviations occur.

Because of this, we have implemented the following two procedures. In one procedure, for the first spot of the reinjection mirror, we calculate the full number of round trips in the absorption cavity. If this reaches a high number, we assume that this will occur for all subsequent spots. As can be seen in Fig. 9, a high spot count gives in general a high enhancement factor. In the second procedure, for all results using the spot count on the reinjection mirror as fitness function in the GA, we always

Fig. 9. Example of the linearity between the number of spots on the reinjection mirror and the scaled enhancement factor using the GA. Parameters are $L = 30$ cm, $r = 99.98\%$, and $R_{1,2,3} = 100$ cm, and x, α_x, α_y, d had optimized values.

verified the enhancement factor (or scaled enhancement factor) by a full calculation using the parameters found at this optimum spot count number. As said before, a full calculation with the GA took 1.2 h for 10 runs, whereas using only the spot count fitness function this reduced to 26 s for 10 runs.

5. CONCLUSIONS

For sensitive laser-based gas spectroscopy, long optical cavities are used to facilitate long interaction path lengths. To perform sensitive gas detection, HR mirrors (>99.95%) are used to achieve the long interaction path lengths. This has the disadvantage that throughput intensity to the detector is low and the sensitivity of the system will be limited by the noise floor of the detector. By using a reinjection mirror before the first cavity mirror, the intensity throughput toward the detector can be increased. To investigate this intensity enhancement, we used a 3D ray tracing model for a three-mirror configuration by applying a grid search and a GA, and taking into account astigmatism, a characteristic often ignored when a ray transfer matrix is used.

Using the grid search, the incoming laser beam angles and cavity parameters are optimized using discrete steps in the parameters. More specifically, we investigated the effect of changing length d of the reinjections' cavity on the enhancement factor, for various absorption cavity characteristics. From these investigations, it is found that a maximum intensity enhancement can be achieved when the d/R ratio is 0.7, irrespective of the mirror radii of the curvatures. Optical cavities in which $L \gg R$ do not give an enhancement, due to the stability criteria of an optical cavity. For confocal absorption cavities ($L = R$) the intensity enhancement is relatively low, while for cavities with ($L = R/2$) better intensity enhancement factors can be found. Using the optimal configurations with maximum enhancement factors, we investigated the tolerances around these optima. It was found that especially the entrance angles' tolerances can be very small, while the position (x) of the entrance hole relative to the central axis and the cavity lengths (d , L) are less critical.

The grid search simulations needed several days, since each of the seven parameters (x , y , α_x , α_y , d , L , $R_{1,2,3}$) had to be varied over its operation range in discrete steps, next to mirror reflectivity. To minimize the calculation time, we investigated the relationship between the enhancement factor and the number of spots on the reinjection mirror.

With the enhancement factor as fitness function, the GA gave strong fluctuating results for the absorption cavity length. Enhancement factors of up to 1400 were found for short cavities (~ 3 cm), and enhancement factors of up to 101 were found for long cavities (50 cm). For short cavities, higher enhancement factors are mainly found due to the astigmatic effect of the short radii of curvature mirrors, combined with larger angles for the incoming beam. As a result, the spot pattern changes from an ellipse into Lissajous figures. When all mirrors have a 2-inch diameter a better enhancement can be found, because the entrance hole diameter becomes smaller compared to the mirror surface. Higher enhancement factors for short cavities mean that the reflectivity of the mirrors can be increased, before the noise limit of the detector becomes a

limiting factor. Longer, effective path lengths can be achieved in short cavities by using higher reflective mirrors with a larger diameter. Furthermore, short lengths of the absorption cavities will open new prospects for compact sensors for ultrasensitive absorption spectroscopy.

The GA approach was not very reproducible when all parameters were free. Because of this and because the cavity length has a direct effect on the gas absorption length, we introduced a new fitness parameter: the scaled enhancement factor. This is the product of the cavity length and the enhancement factor. Using this parameter as fitness parameter the GA became more reproducible.

In conclusion, we found that the GA is an order of magnitude faster in calculation time for finding optimal solutions, using the parameters as continuous functions. However, combining the GA with the grid search will give a more fruitful outcome: the GA generates a maximum result, while the grid search calculates the parameter tolerance around these optimal solutions.

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