Numerical analysis of beam propagation in pulsed cavity ring-down spectroscopy

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A numerical simulation of pulsed cavity ring-down spectroscopy (CRDS) is developed with the commercially available software package GENERAL LASER ANALYSIS AND DESIGN. The model is verified through a series of numerical experiments. Several issues of concern in CRDS are investigated, including spatial resolution, misalignment, non-Gaussian beam input, and the effect of flames inside a ring-down cavity. Suggestions for the design of pulsed CRDS instruments are provided. © 2002 Optical Society of America

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1. Introduction

Cavity ring-down spectroscopy (CRDS) is a direct absorption technique based on the measurement of the decay rate of a laser beam trapped in a high-finesse optical resonator that makes multiple passes through an absorber. Early CRDS systems used pulsed laser sources. Improvements in spectral resolution, data acquisition rates, and sensitivity have been made through the use of narrow-linewidth continuous-wave (cw) lasers. Despite these improvements, pulsed CRDS continues to be useful owing to its simplicity and to the reliability, tuning range, and output power provided by pulsed laser systems. Pulsed CRDS has been an especially useful diagnostic for absolute species measurements in combustion flows.

An analytical electromagnetic analysis of CRDS was performed by Hodges et al. in 1996. That analysis provided a detailed description of modes inside pulsed CRDS resonators. Although we emphasize different aspects, our model is not in disagreement with Hodges et al. In this paper we present results of a numerical analysis of pulsed CRDS that describe beams inside CRDS resonators. Our simulations were performed with GENERAL LASER ANALYSIS AND DESIGN (GLAD), a physical optics code that numerically propagates an electromagnetic wave. The code provides solutions to the full diffraction problem. GLAD uses computational utilities that can be linked via calls (a simplified, specialized computer language) and includes postprocessing and image presentation.

GLAD solves the diffraction equation given by

\[ \frac{\partial \hat{A}}{\partial z} = -i \frac{1}{2k} \nabla \cdot \hat{A} - i \frac{\mu \omega^2}{2k} \hat{P}, \]

where

\( \hat{A} \) is the complex amplitude of the wave, given here by \( |\hat{A}| = \sqrt{c \varepsilon / 2n |E_0|} \) [then irradiance \( I = A^2 = (A \ast A) \)];

\( c \) is the speed of light in vacuum;

\( \varepsilon \) is the permittivity in the medium;

\( n \) is the refractive index of the medium;

\( E_0 \) is the complex electric field amplitude;

\( z \) is the direction of propagation of the wave;

\( i \) is the \( \sqrt{-1} \);

\( k \) is the magnitude of the wave vector (\( k = 2\pi / \lambda \));

\( \nabla \cdot \) is the transverse Laplacian operator (neglecting the \( z \)-dependent terms);

\( \mu \) is the magnetic permeability in the medium;

\( \omega \) is the radial frequency (\( = 2\pi c / \lambda \));

\( \hat{P} \) is the material polarization.

The development of Eq. (1) assumes that the field is a harmonic wave (\( \hat{A} \) is written as a complex quantity, in terms of an amplitude and phase). Moreover, we assume that changes with \( z \) are negligible relative to...
changes in the transverse direction and that $\tilde{A}$ varies slowly with $z$ (allowing one to neglect second derivatives with respect to $z$).

GLAD solves Eq. (1) by use of a split-step method, in which diffraction and interaction with the medium are separated into two steps:

\[
\Delta A_{\text{diffraction}} = -i \frac{1}{2k} \nabla^2 A \Delta z, \tag{2}
\]

\[
\Delta A_{\text{medium}} = -i \frac{\mu_0 w^2}{2k} P \Delta z = -i \frac{k \chi}{2n^2} A \Delta z, \tag{3}
\]

where $P$ has been written in terms of the material susceptibility $\chi$. The vector notation of $A$ has been removed because GLAD operators explicitly determine the polarization for their own problems. If suitably small step sizes are chosen, one can numerically solve Eq. (2) first, then Eq. (3). The material interaction portion [Eq. (3)] requires a straightforward numerical integration. The diffraction portion [Eq. (2)], which is somewhat more complex, is solved with a fast-Fourier-transform (FFT)-based approach. In the FFT approach, it is necessary to determine whether the problem (at a specific location) is in the near or the far field. This question is answered by propagation of a surrogate Gaussian beam along with the numerical solution. The surrogate beam is propagated by use of the standard Gaussian beam formulas that are written in terms of irradiance. By monitoring of the surrogate beam size and phase radius of curvature, GLAD makes changes to the numerical approach to optimize accuracy. Significantly more detail is provided by Lawrence.\(^{10}\)

Several issues are addressed by use of the model, including spatial resolution, misalignment, non-Gaussian beam input, and the effect of flames inside a ring-down cavity. Each researcher, however, has specific goals, requirements, and constraints for their own CRDS experiment. The data are therefore presented in a nondimensional format, allowing readers to adopt our model results to their own cases and to make their own judgments regarding what is and is not acceptable performance.

### A. Definition of Nondimensional Cavity Parameters

To begin, we provide a brief review of resonator theory and notation. A more detailed review can be found in Siegman.\(^{11}\) To present our model results, we use the resonator $g$ parameter, defined as

\[
g_1 = 1 - \frac{L}{R_1}, \quad g_2 = 1 - \frac{L}{R_2}, \tag{4}
\]

where $L$ is the length of the cavity and $R$ is the mirror radius of curvature. The $g$ parameters are confined to a stability range defined by $0 \leq g_1 g_2 \leq 1$ as shown in Fig. 1.

If the point $g_1 g_2$ falls in unstable regions outside the shaded area, no Gaussian beam can be sustained between the mirrors in a periodic focusing system.

The most commonly used resonator for pulsed CRDS consists of two mirrors with equal radii of curvature ($R_1 = R_2$) so that $g = 1 - L/R$. This stability region shown in Fig. 1 is the line that lies along the $g_1 = g_2$ plane from $g = 1$ (planar) through $g = 0$ (confocal) to $g = -1$ (concentric). It is important to note that these three cavity geometries, planar, confocal, and concentric, all lie on the stability boundary and are therefore prone to instability.

For a stable symmetric resonator, the beam size (or Gaussian spot) of the lowest-order transverse mode of the cavity is defined as

\[
w_0^2 = \frac{L \lambda}{\pi} \left[ \frac{1 + g}{4(1 - g)} \right]^{1/2},
\]

\[
w_1^2 = w_2^2 = \frac{L \lambda}{\pi} \left( \frac{1}{1 - g^2} \right)^{1/2}, \tag{5}
\]

where $w_0$ is the beam size in the center of the cavity and $w_1 = w_2$ are the beam sizes at the cavity mirrors. In this paper we report the beam size in the nondimensional form: $(\pi w^2/L\lambda)^{1/2}$.

To couple properly into this lowest-loss transverse mode of the cavity, it is necessary to shape and focus (or mode match) the input beam. We discuss and model the effects of a beam that is not properly aligned and mode matched in a later section of the paper. It is clear, however, from Eqs. (5) that shorter cavity lengths reduce the beam size inside the resonator. This is an important factor to consider when spatial resolution is a concern (e.g., CRDS measurements in nonhomogeneous media). The beam size at the cavity center and at the mirrors is shown in Fig. 2 (same as Fig. 19.6 of Siegman\(^{11}\)) for the entire range of stability. From the graph, one can

![Stability diagram for a two-mirror optical resonator](adapted from Fig. 19.4 of Siegman\(^{11}\)).
The decay rate in CRDS is a function of the finite-diameter resonator-mode losses exactly with the theoretical results shown in Fig. 2. The mirrors was calculated. The model results agree 0.95 launched into a resonator an exactly mode-matched Gaussian beam was

In an effort to verify the model, we performed several numerical experiments including measurement of the finite-diameter resonator-mode losses and transverse-mode frequencies. For initial verification, an exactly mode-matched Gaussian beam was launched into a resonator (ranging from \(-0.95 \leq g \leq 0.95\)), and the beam size at the cavity center and at the mirrors was calculated. The model results agree exactly with the theoretical results shown in Fig. 2.

### 1. Finite-Diameter Resonator-Mode Losses

The decay rate in CRDS is a function of the finite reflectivity of the end mirrors, absorption and scattering of the medium inside the resonator, and transverse-mode-dependent diffraction losses at the mirrors. An important feature of the model is the ability to monitor these diffraction losses.

In our simulations we model the losses from an exactly mode-matched Gaussian beam while varying the diameter of the resonator mirrors. Figure 3 shows the model results of the power loss per one-way pass (in decibels) versus the Fresnel number \(N_f\), which is an indicator of how large the resonator aperture is compared with the mode size, defined as

\[
N_f = \frac{a^2}{L\lambda},
\]

where \(2a\) represents the transverse width of the resonator end mirrors. The cavity loss that was due to a finite mirror reflectivity of 99.99% is included on the plot for reference. The model diffraction losses agree with published theoretical results (same as Fig. 19.20 of Siegman).

### 2. Transverse-Mode Frequency

The analysis by Hodges \(9\) demonstrated that longitudinal modes should be included in a complete model for CRDS behavior. In this study we focus exclusively on the spatial-mode properties of pulsed CRDS cavities. To describe spatial behavior does not require a simultaneous description of longitudinal modes. Longitudinal modes can be described in GLAD, but the approach taken by Hodges \(9\) is perhaps more appropriate for that task. The laser pulse launched into a cavity often excites higher-order transverse modes. To validate the presence of these higher-order modes in the modeled cavities, we designed a numerical experiment to measure the transverse-mode beat frequency.

As a brief review, the resonance frequencies of the

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**Table 1. Cavity Geometry Utilized by the Pulsed CRDS Community**

<table>
<thead>
<tr>
<th>Researcher(s)</th>
<th>(R) (cm)</th>
<th>(L) (cm)</th>
<th>(g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jongma et al.(12)</td>
<td>25</td>
<td>45</td>
<td>-0.8</td>
</tr>
<tr>
<td>Spanjaars et al.(13)</td>
<td>25</td>
<td>41.2</td>
<td>-0.648</td>
</tr>
<tr>
<td>Mercier et al.(6)</td>
<td>25</td>
<td>40</td>
<td>-0.6</td>
</tr>
<tr>
<td>Mercier et al.(14)</td>
<td>25</td>
<td>40</td>
<td>-0.6</td>
</tr>
<tr>
<td>Derzy et al.(15)</td>
<td>50</td>
<td>75</td>
<td>-0.5</td>
</tr>
<tr>
<td>Lozovsky and Cheskis(16)</td>
<td>50</td>
<td>75</td>
<td>-0.5</td>
</tr>
<tr>
<td>Romanini and Lehmann(2)</td>
<td>100</td>
<td>130</td>
<td>-0.3</td>
</tr>
<tr>
<td>Evertsen et al.(5)</td>
<td>25</td>
<td>30</td>
<td>-0.2</td>
</tr>
<tr>
<td>Hodges et al.(9)</td>
<td>100</td>
<td>118</td>
<td>-0.18</td>
</tr>
<tr>
<td>Meijer et al.(9)</td>
<td>10</td>
<td>11.5</td>
<td>-0.15</td>
</tr>
<tr>
<td>Sappey et al.(17)</td>
<td>100</td>
<td>98</td>
<td>0.02</td>
</tr>
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<td>Bogaards and Meijer(18)</td>
<td>25</td>
<td>18</td>
<td>0.28</td>
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<td>Zalicki and Zare(19)</td>
<td>200</td>
<td>106</td>
<td>0.47</td>
</tr>
<tr>
<td>van Zee et al.(20)</td>
<td>20</td>
<td>10.5</td>
<td>0.475</td>
</tr>
<tr>
<td>Xie et al.(21)</td>
<td>100</td>
<td>41.5</td>
<td>0.585</td>
</tr>
<tr>
<td>Martin et al.(22)</td>
<td>200</td>
<td>41</td>
<td>0.795</td>
</tr>
<tr>
<td>Thoman and McIlroy(7)</td>
<td>600</td>
<td>72.5</td>
<td>0.879</td>
</tr>
<tr>
<td>Paul and Saykally(23)</td>
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<td>0.917</td>
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<tr>
<td>McIlroy(24)</td>
<td>600</td>
<td>39</td>
<td>0.935</td>
</tr>
<tr>
<td>Dreyer et al.(8)</td>
<td>400</td>
<td>21.5</td>
<td>0.946</td>
</tr>
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</table>
axial-plus-transverse modes in a cavity are described in Siegman11 as

\[ q + (n + m + 1) \frac{\cos^{-1}(g)}{\pi} \frac{c}{2L}, \]  

(7)

where \( c \) is the speed of light, \( q \) is the axial mode, and \( m \) and \( n \) are the even and odd transverse modes.

It is evident from this equation that the transverse modes are degenerate when \( \frac{\cos^{-1}(g)}{\pi} \) is an integer.

The most common degenerate cavity is the confocal resonator, \( g = 0 \), also referred to as a magic number 2 cavity because the transverse modes are separated by one half the longitudinal mode spacing given by \( c/2L \).

To verify transverse-mode beating in our model, we launched a beam with a top-hat profile into a degenerate geometry cavity. The intensity profile of the beam propagating inside the cavity was highly structured and varied from pass to pass, indicating the excitation of relatively high-order transverse modes.

In our model the beam energy escaping from this ring-down cavity was partially blocked with a rectangular obscuration. The obscuration is representative of the strip of paper placed in front of the detector in the experiments of Hodges et al.9 A discrete FFT was performed on this decay waveform to determine the transverse-mode beat frequency. The numerical model results for a mirror radius of curvature equal to 1 m are shown in Table 2. The table lists the cavity length, \( L \), the corresponding \( g \) parameter, the magic number, theoretical frequencies, experimental results published by Hodges et al.,9 and our model results for the identical cavities. Figure 4 shows the FFT of a simulated ring-down signal for a magic number 4 degenerate cavity. The transverse-mode beat frequency of 128 MHz \((c/8L)\) is clearly seen.

### Table 2. Transverse-Mode Frequency for Degenerate Cavities

<table>
<thead>
<tr>
<th>( L ) (cm)</th>
<th>( g )</th>
<th>Magic Number</th>
<th>Theory (MHz)</th>
<th>Hodges et al.9 (MHz)</th>
<th>GLAD model (MHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.0</td>
<td>2</td>
<td>75</td>
<td>75</td>
<td>75</td>
</tr>
<tr>
<td>50</td>
<td>0.50</td>
<td>3</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>29.289</td>
<td>0.71</td>
<td>4</td>
<td>128</td>
<td>128</td>
<td>128</td>
</tr>
<tr>
<td>19.098</td>
<td>0.81</td>
<td>5</td>
<td>157</td>
<td>157</td>
<td>157</td>
</tr>
<tr>
<td>13.39</td>
<td>0.87</td>
<td>6</td>
<td>187</td>
<td>187</td>
<td>187</td>
</tr>
</tbody>
</table>

3. Ring-Down Waveforms

For a final test we investigated the time-domain behavior of the model for an empty cavity and with absorption losses. We modeled our numerical experiment after that of Zalicki et al.25 The prediction was based on a laser bandwidth that is a delta function relative to the absorption linewidth, tuned to line center. Although that is a simplified result that can be modeled with standard CRDS formulas, it does test whether GLAD produces reasonable temporal decays. Figure 5 shows the simulated ring-down waveforms (refer to Fig. 2 in Zalicki et al.25 for comparison).

The upper and lower waveforms shown in the figure are the model results when the absorbance in the cavity was zero and 0.0012, respectively. A first-order exponential decay curve was fitted to the simulated data and ring-down times were calculated to be approximately 195 ns (on resonance) and 172 ns.
The model results agree with CRDS theory and the experimental results reported by Zalicki et al.\textsuperscript{25} The results of these numerical experiments confirm that GLAD is capable of modeling the details of CRDS behavior.

2. Types of Beam Used as Model Input

The lowest-order transverse-mode of a stable resonator has a Gaussian transverse profile (TEM\textsubscript{00}). These Gaussian beams are widely used in the analysis of laser beams and related optical systems, and therefore we begin our modeling with this type of beam. However, the output beams of most pulsed laser systems do not have a Gaussian transverse profile because the pulsed laser resonator is unstable by design. In an effort to model a beam more like the output of a pulsed laser, super-Gaussian beams were investigated with a transverse profile of the form

\[ E(r) = \exp \left[ -\left( \frac{r^2}{w^2} \right)^n \right], \quad n \geq 2, \quad (8) \]

where \( r \) is the transverse coordinate, \( w \) is the Gaussian transverse radius, and \( n \) is the super-Gaussian exponent. A beam with a super-Gaussian exponent equal to 3 was selected for our model as this is a close match to the spatial beam profile of our pulsed laser system. Figure 6 shows cross-sectional plots for these two beam types.

3. Model Results

A. Mode-Mismatched Cavities

Exact mode matching is necessary to create a standing wave in which the beam waist resides at the cavity center over the lifetime of the ringdown. The researcher attempting to mode match a beam into a cavity must control two beam parameters: the phase radius of curvature and the beam size. In practice, this is a challenging task. If the input beam is not properly aligned and mode matched to the transverse pattern of the lowest-order mode, the input wave will excite some mixture of lowest-order and higher-order transverse modes in the cavity. The mode-mismatched condition was simulated, starting with Gaussian beam input. Figure 7 shows the beam profile for an exact mode-matched condition and two mismatched cases.

Figure 7(a) shows the results for an exactly mode-matched input beam. This is a standing wave in which the beam waist sits at the cavity center and is larger at the mirrors and the pattern repeats indefinitely. In Fig. 7(b) the input-beam radius is 50\% larger than the exact mode-matched size [as calculated by Eqs. (5)] and the phase radius of curvature is exact. As seen in Fig. 7(b), the beam size is smaller in the middle of the cavity versus at the mirror; however, it increases after each pass in the resonator. The beam waist moves slightly from the cavity center for this case. If the simulation is run long enough, the pattern will repeat. The time it takes for this beam-size cycle to repeat, or its breathing frequency, is not the same as the transverse-mode beat frequency discussed previously. In Fig. 7(c) the input beam has a phase radius of curvature 25\% greater than an exact mode match (the mode-matched phase radius of curvature is equal to \( -R \)), and the beam size...
is exact. In this case the beam waist clearly moves off the cavity center. The phase-mismatched beam will also repeat if the simulation is run long enough. It is clear from Fig. 7 that a mismatched beam degrades spatial resolution in CRDS. In the two mismatched cases, placing an aperture at the resonator output will increase the noise level in the signal if the aperture is in between the smallest and the largest beam sizes. It is important to note here that the change in the ring-down time is negligible owing to mismatching of this magnitude in an empty resonator.

Because the mismatched input excites higher-order modes in the cavity, the beam size at the center of the resonator varies in time and is effectively larger than Eqs. (5) predict. In an effort to aid researchers attempting to optimize spatial resolution of CRDS, we investigated the effective beam size at the cavity center as a function of the $g$ parameter. Figure 8 shows the effective beam size at the cavity center versus $g$ when the beam size is mismatched (input-beam size 50% larger than the exact mode-matched size and the phase radius of curvature is exact). Figure 9 shows the model results where the phase radius of curvature is mismatched 25% and the beam size is exact.

The shaded regions in these graphs represent the limits of the beam size at the center of the cavity. The dark gray curves shown in both graphs labeled "exact mode match" is the same line labeled $w_0$ in Fig.
modes is discussed in more detail by Meijer et al.\textsuperscript{3} and Hodges et al.\textsuperscript{9} Fortunately, degenerate cavities are easy to avoid.

Super-Gaussian beam input, by definition, cannot match the transverse pattern of the lowest-order cavity mode. Even when the beam size and phase radius of curvature are exactly matched to the lowest-order transverse-mode of the cavity, they will still excite a mixture of lowest-order and higher-order transverse modes in the resonator. Figure 10 shows model results for the beam size at the cavity center as a function of \( g \) in which the super-Gaussian input is exactly mode matched as if it were a true Gaussian beam.

The notches at the degenerate cavity locations are seen again in this case, as expected. What may be surprising is the magnitude of size range. It is only slightly less than in the case in which a Gaussian beam was size mismatched \( 50\% \). The point is that even if painstaking effort is taken to mode match a beam to a CRDS resonator, the spatial resolution as determined by Eqs. (5) will be largely overestimated if the input beam is not Gaussian.

Because the beam profile from most pulsed laser systems is not TEM\(_{00}\), researchers often spatially filter the beam through a circular aperture to form a quasi-Gaussian transverse profile. We investigated the effect of spatially filtering our super-Gaussian beam. A collimated super-Gaussian beam was propagated through a 25-mm focal-length lens, with an aperture placed at the focus, and another 25-mm focal-length lens used to recollimate the beam. The beam size at the cavity center was reduced \( 10\% \) (versus without spatial filtering) when the diameter of the aperture was 1.5 times the focus spot size.

There are some comments that can be made regarding the sensitivity to the mode matching in general. Mode-match sensitivity for mismatched input size and the super-Gaussian beam is minimized as \( g \rightarrow -1 \), whereas the phase radius of curvature mismatch is minimized as \( g \rightarrow 1 \).

B. Cavity Misalignment

We are able to misalign our model cavity in several ways: mirror tilt, mirror centers offset from one another, input beam offset from the cavity center, and input-beam angle. When a ring-down cavity is slightly misaligned, higher-order transverse modes are excited, which results in larger beam sizes, and the spatial resolution of the experiment is further compromised by beam displacement. Figure 11 shows the beam size and location for aligned and slightly misaligned resonators. The simulation was run for 0.5 \( \mu \)s, and the beam size and position were calculated at the output mirror—much like a CCD camera placed at the output mirror would record. In the figure the beam centers are shown as points in \( x\text{-}y \) space while the beam size is represented by \( x\text{-}y \) error bars. In the aligned case, the beam remains at the same point on the \( x\text{-}y \) axis; however, the beam size does change (a super-Gaussian input beam was used), giving the appearance of a thick error bar cap. In the misaligned case, one of the resonator mirrors was tilted 0.005° (horizontally), and the beam was input at a 0.001° angle in the horizontal plane. In the misaligned case, the beam is initially displaced and shown to “walk” back and forth over time. This numerical experiment was modeled after the re-
search of Thoman and McIlroy\textsuperscript{7} in which camera images from both aligned and misaligned cavities were reported. Their experimental images are similar to our numerical results.

To judge the seriousness of misalignment effects, we modeled two typical cavity misalignments as a function of $g$. Figure 12 shows the beam displacement calculated at the cavity mirror for a 1-mm input-beam offset and a 0.5-deg mirror tilt per unit cavity length (at one mirror only). The model results show that displacement is relatively insensitive to an offset input beam when $g > 0$ and increasingly sensitive as $g$ approaches $-1$. Sensitivity to mirror tilt is a function of the cavity length and $g$. The shorter the cavity, the less sensitive it is to tilt misalignment. Beam displacement that is due to mirror tilt is a function of the cavity length and $g$. The shorter the cavity, the less sensitive it is to tilt misalignment.

C. Effect of Flames in a Ring-Down Cavity

1. Atmospheric Pressure, Postflame

Combustion researchers have speculated what effect a flame has in a CRDS resonator; however, the extent to which the beam is perturbed, if at all, has yet to be answered. One would expect that a flame would act like a weak diverging lens as the increased temperature gradient results in a lower index of refraction for air. Figure 13 shows a experimental temperature profile in the postflame region of a 2-cm-diameter symmetric atmospheric flat-flame burner (burner details can be found in Dreyer \textit{et al.}\textsuperscript{8}) and the corresponding index of refraction of air calculated via the Edlen empirical formulas\textsuperscript{26}.

We have studied beams near the center of the flame because CRDS is not spatially resolved. It therefore will be used primarily in uniform flames. In such a case, CRDS beams will most commonly be directed down the center of the flame where the absorption path length is easy to measure and where possible lensing effects will be minimized.

We began our analysis of flames in CRDS resonators by approximating the flame index change as a spherical thin lens. Utilizing the standard thin-lens equation (also known as the lens maker's formula), one can estimate the focal length of the flame–lens. For the atmospheric-pressure postflame experimental data shown above, we assumed a lens surface radius of curvature of 1.5 cm and an index change of $2.3 \times 10^{-4}$ (1850 K at atmospheric pressure), which corresponds to a lens with an approximate focal length of $-32.5$ m. A thin lens with this focal length was inserted at the cavity center of our model.

It is important to note that the model results did not indicate any beam steering or deflection when the beam passed through the center of the lens or with the flame offset as much as 1 mm. The beam size was affected by the presence of the lens, not as a function of the $g$ parameter, but as a function of the mirror radius of curvature, $R$. Figure 14 shows the percent increase in beam size as a function of mirror radius of curvature when a thin lens described above was inserted at the cavity center. Model results indicate that large mirror radii of curvature exacerbate the divergence effect of the lens, which is formed by the temperature gradient in the flame. A reason-
able conclusion would be to use mirrors with smaller radii of curvature when one is optimizing the spatial resolution of a CRDS experiment.

Adding a layer of complexity to the simulation, we approximated the flame index change as a thick lens constructed from the experimental flame-temperature data in Fig. 13. The center portion of the flame, at which the temperature and index are flat, was modeled as a spherical region of low index surrounded by two thin boundary regions in which the indices of refraction are between that of the flame center and the surrounding air. Construction details of the thick lens are shown in Figure 15.

The focal length of this thick lens was calculated to be \(-30.9\) m. Mode-matched Gaussian and super-Gaussian beams were launched into a simulated CRDS resonator with this thick lens at the cavity center. The results are shown in Fig. 16.

The thick-lens model of an atmospheric-pressure postflame showed only minor perturbation of the Gaussian beam input. As in the thin-lens case, the beam was not measurably displaced (steered) in the cavity when the lens was present, and the beam size showed a strong correlation to the mirror radius of curvature, \(R\). The largest beam-size increase occurs as \(R\) approaches 400 cm at \(g = 0.95\). As shown in the graph, there is a region of increased beam-size deviation centered around the confocal cavity geometry and, to a lesser extent, the degenerate \(g = \pm 0.5\) geometry. The increased size at the confocal region is due to its inherent instability (refer to the stability diagram, Fig. 1.) The super-Gaussian beam input was perturbed by the flame slightly more (approximately 0.5%) than the Gaussian input in general. The results show a similar instability at degenerate geometries and the increased divergence when larger mirror radius curvature is used.

2. Other Flame Types

Combustion researchers are often interested in studying the flame-front region. In that light, we investigated a 2-cm-diameter atmospheric-pressure Bunsen flame, with a beam passing through the flame front, at the edge of the cone. The flame was modeled as a 2-cm-diameter, 50-\(\mu\)m-thick spherical low-index region representative of the flame front at
2000 K. Inside the flame front the index of refraction was equivalent to 500 K air. Model results indicate the beam is steered slightly, approximately 0.1 mm on the mirror. The beam energy, however, is completely attenuated by diffraction losses at the mirrors after approximately ten round-trip passes in the cavity. The beam position on the mirror oscillates in a composite pattern that is large enough to emulate a low Fresnel number with high loss, as shown in Fig. 3. Atmospheric-pressure flames impose a difficult constraint on the CRDS beam for this reason. Unless the beam is steered slightly, approximately 0.1 mm, the CRDS intracavity beam will be too large to resolve the structure of an atmospheric-pressure flame front. It is our recommendation, therefore, that CRDS investigations of flame-front chemistry be performed in low-pressure flames.

CRDS measurements in a flame front are usually performed at low pressure, around 50 to 200 mbar, to expand the reaction zone with a flat-flame burner. At a pressure of 100 mbar, the index change owing to temperature is roughly an order of magnitude less than at atmospheric pressure. Therefore, at low pressure, a beam passing through the center of a postflame region similar to the Meeker burner modeled above would be disturbed even less. Another factor that minimizes the effect of the flame is burner diameter. For example, the approximate focal length for a 6-cm-diameter flame at 1850 K would be approximately \(-528 \text{ m}\) at a low pressure of 100 mbar. These two factors minimize beam perturbation in a low-pressure flame.

Recently, McIlroy\(^ {27}\) reported measuring a small baseline shift in CRDS spectra with vertical displacement through the flame zone of low-pressure flames. We have reported similar baseline shifts in our CRDS low-pressure flame research.\(^ {28}\) We can find no loss mechanism in our model that explains this phenomenon other than particulate scattering. This is a reasonable conclusion as CRDS has been proven to be extremely sensitive to scattering from small particulates (Sappey et al.\(^ {17}\)).

4. Conclusions and Recommendation

We have demonstrated the ability to numerically model beam propagation in a pulsed CRDS resonator, investigating several issues such as spatial resolution, misalignment, super-Gaussian beam input, and the effect of flames. Some general conclusions can be distilled from these simulation results. One should use the shortest cavity length, \(L\), feasible to optimize spatial resolution. Non-Gaussian input and nonexact mode matching degrade the spatial resolution of CRDS. Therefore careful mode matching and spatial filtering should be practiced when one optimizes this parameter. Confocal geometries should be avoided when large temperature gradients are present inside a CRDS resonator. For pulsed CRDS, degenerate geometries in general should be avoided because of the large mode spacing inside the resonator. Extreme near-planar and near-concentric cavity geometries should be avoided as they have been shown to be more sensitive to misalignment and mode-matching precision. A resonator with a \(g\) parameter in the range of 0.5 appears to be the least sensitive to the types of misalignment modeled herein. Beam perturbation from flat atmospheric postflame gases was found to be minimal; however, large mirror radii of curvature exacerbate the divergence effect of the flame. Larger diameter burners and lower pressures should further reduce the weak lensing effect of flames. Beam steering in atmospheric-pressure flame fronts can have a significant effect on CRDS signals.

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References


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