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Comparison of line-peak and line-scanning excitation in two-color laser-induced-fluorescence thermometry of OH

Stanislav Kostka  
*University of Connecticut - Storrs*

Sukesh Roy  
*Spectral Energies*

Patrick J. Lakusta  
*Air Force Institute of Technology*

Terrence R. Meyer  
*Iowa State University, trm@iastate.edu*

Michael W. Renfro  
*University of Connecticut - Storrs*

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Abstract

Two-line laser-induced-fluorescence (LIF) thermometry is commonly employed to generate instantaneous planar maps of temperature in unsteady flames. The use of line scanning to extract the ratio of integrated intensities is less common because it precludes instantaneous measurements. Recent advances in the energy output of high-speed, ultraviolet, optical parameter oscillators have made possible the rapid scanning of molecular rovibrational transitions and, hence, the potential to extract information on gas-phase temperatures. In the current study, two-line OH LIF thermometry is performed in a well-calibrated reacting flow for the purpose of comparing the relative accuracy of various line-pair selections from the literature and quantifying the differences between peak-intensity and spectrally integrated line ratios. Investigated are the effects of collisional quenching, laser absorption, and the integration width for partial scanning of closely spaced lines on the measured temperatures. Data from excitation scans are compared with theoretical line shapes, and experimentally derived temperatures are compared with numerical predictions that were previously validated using coherent anti-Stokes–Raman scattering. Ratios of four pairs of transitions in the $A^{2}\Sigma^{+}\leftrightarrow X^{2}\Pi (1,0)$ band of OH are collected in an atmospheric-pressure, near-adiabatic hydrogen-air flame over a wide range of equivalence ratios—from 0.4 to 1.4. It is observed that measured temperatures based on the ratio of $Q_{1}(14)/Q_{1}(5)$ transition lines result in the best accuracy and that line scanning improves the measurement accuracy by as much as threefold at low-equivalence ratio, low-temperature conditions. These results provide a comprehensive analysis of the procedures required to ensure accurate two-line LIF measurements in reacting flows over a wide range of conditions.

Keywords
cohesent scattering, fluid structure interaction, fluorescence, gas absorption, laser produced plasmas, microlenses, scanning, temperature sensors, coherent anti-Stokes, collisional quenching, comprehensive analysis, energy output, equivalence ratios, gas phase, laser absorption, laser induced fluorescence, LIF thermometry, low temperatures, measurement accuracy, numerical predictions

Disciplines
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Comments

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1Department of Mechanical Engineering, University of Connecticut, 191 Auditorium Road, U-3139, Storrs, Connecticut 06269, USA
2Spectral Energies, LLC, 5100 Springfield Street, Suite 301, Dayton, Ohio 45431, USA
3Department of Aeronautics and Astronautics, Air Force Institute of Technology, 4950 Hobson Way, Wright–Patterson Air Force Base, Ohio 45433, USA
4Department of Mechanical Engineering, Iowa State University, 3034 Black Engineering Building, Ames, Iowa 50011, USA
5Air Force Research Laboratory, Propulsion Directorate, Wright–Patterson Air Force Base, Ohio 45433, USA
*Corresponding author: sroy@woh.rr.com

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1. Introduction

Combustion research relies on temperature measurements to aid the understanding of chemical interactions in complex flows; such research may
involve determining the operating conditions for an advanced combustor or developing and validating computer models. Over the past few decades, several nonintrusive laser diagnostic techniques, such as Rayleigh scattering, Raman scattering, coherent anti-Stokes–Raman scattering (CARS), laser-induced fluorescence (LIF), and absorption spectroscopy, have been used for measuring temperature by a major species, for instance, \( \text{N}_2 \). The oscil- lating polarization of the molecule is coupled with the vibrational frequencies to scatter light at wavelengths that are shifted by the vibrational energy of the molecule. A spectral fit is applied to the acquired vibrational spectrum for extracting the gas-phase temperature. Another technique used for ther- mometry is CARS, which involves a coherent beam that is generated within the measurement volume where the entire signal can be collected with good rejection of background light. CARS measurements have been performed since the 1970s; a review of work done by Eckbreth can be found in Ref. [1] and an overview of the technique presented by Hall and Eckbreth in Ref. [5]. More recent work performed by Roy et al. [6] has shown that, when this technique is applied to diamond-forming flames, it provides temperature accuracies of about \( \pm 4\% \). Hancock et al. [7] applied this technique to a Hencken burner similar to the one used for the present study and reported temperature accuracies of \( \sim 3\% \). Despite its accuracy, the CARS technique has some disadvantages. The first is the complexity of the experimental setup, which involves the crossing of multiple laser beams and maintenance of spatial overlap or phase matching between those beams in harsh operating conditions. Analysis of the spectroscopic data is also cumbersome, requiring precise modeling of the nonlinear behavior. As opposed to CARS, LIF is a linear technique and its implementation more straightforward since crossed beams are not required. LIF can also be readily configured for point, line, planar, or even volume measurements.

The goals of the present study were to examine the accuracy of ratio-of-line-center versus ratio-of-line-scanning LIF-based techniques for temperature measurements in flames and to assess the optimum application of this technique for various OH transitions. One approach discussed extensively in the liter- ature for determining the temperature-dependent Boltzmann distribution of the population is the measurement of two different OH transitions at line center. Another thermometry approach is the two-line scanning method, in which the temperature can be determined from the ratio of the integrated line intensities of two OH transitions; this technique is seldom used because of its limitations with regard to unsteady/turbulent flames and longer collection times, when compared to line-peak thermometry. However, recent work by Miller et al. [8] has shown that rapid scanning of OH line shapes for two-line planar thermometry is possible; it was of interest in the present study to determine whether line scanning would offer a significant improvement in temperature accuracy over the measurement of line-center intensity ratios. This includes analysis of partial line scanning over two closely spaced OH transitions. Through simulation of line shapes, the area ratio of various transitions with respect to tem- perature can be predicted and these ratios then used to extract temperature from experimental data. A two-line scanning technique has been used with diode-laser absorption by Liu and co-workers, who employed both direct absorption and 2f wavelength-modulation scans [9]. LIF thermometry using OH was originally performed by Cattolica [10]. Since then, several reviews of thermometry involving this technique have appeared in the literature [1,3,4,11]. Laurendeau [4] has reviewed LIF thermometry that employs excitation scans, fluorescence scans, and two-line fluorescence, including a discussion of two- line saturated fluorescence [12]. The present paper considers only measurements within the linear re- gime of fluorescence.

In the present research effort, various transition pairs commonly used for thermometry were investig- ated to determine the best line pair for measuring gas-phase temperature based on adiabatic flame temperatures previously verified using CARS. Accu- racy of experimentally determined temperatures to calculated temperatures using a line-area ratio was compared with the line-peak ratio. The line pair yielding the best results when compared to theo- retical temperatures using the line-peak-ratio meth- od was used for this comparison. The advantages and disadvantages of the peak-intensity and integrated-intensity methods were evaluated and are discussed herein.

Although significant work has been accomplished using various LIF thermometry techniques, reports of studies comparing the accuracy of the two-line thermometry techniques are limited. Some research- ers, notably Seitzman and co-workers [13,14], have developed rules governing the selection of these
lines. Hanson proposed two major rules for the selection of transition pairs—(1) only those transitions with \(J\) between 5 and 12 should be used since the signal from \(J = 12\) maintains an adequate signal level for temperatures between 1000 and 3000 K, and (2) only those transitions isolated from other nearby lines should be used [13,14]. Seitzman et al. tested three different line pairs—the \(Q_2(11)\) paired with \(R_2(8), P_1(7),\) and \(R_2(5)—\) and found that the \(P_1(7)/Q_2(11)\) and \(R_2(8)/Q_2(11)\) transition pairs yielded higher accuracy than the \(R_2(5)/Q_2(11)\) pair within the range of operating conditions examined [14].

While Seitzman et al. showed the \(P_1(7)/Q_2(11)\) ro-vibrational line pair to be suitable for thermometry, others continued to use different line pairs. Welle et al. [15] used the \(Q_1(5)/Q_2(11)\) line pair for saturated LIF thermometry in a propane/air counterflow diffusion flame. Giezendanner-Thoben et al. [16] employed the \(P_1(2)/R_2(13)\) line pair for temperature measurements within a pulsating-gas-turbine model combustor where the temperature accuracy was determined to be 4%–7%, despite the fact that both transitions were outside the limit suggested by Seitzman et al. [14]. The present paper will use the line pairs described above employing the line-peak ratio, followed by their use with a line-area ratio for a line pair that produces the best fit to the theoretical temperatures. The effects of absorption and spectral-overlap corrections on the measured temperature for the peak-intensity ratio technique are discussed.

In addition to comparing the accuracy of the two OH thermometry techniques, this paper also addresses the importance of the selection of isolated line pairs. The \(Q_1(8)/Q_2(9)\) line pair will be added to the previously mentioned line pairs for the purpose of examining the temperature accuracy when transition lines are in close proximity.

2. Experimental Setup

Fluorescence measurements were conducted above the surface of a near-adiabatic Hencken burner, shown in Fig. 1, which is similar to that used by Woolridge et al. [17], Kulatiilaka et al. [18], Hancock et al. [7], and Meyer et al. [19]. The burner consists of a 24 mm square, Hastalloy honeycomb that supports hypodermic needles within a quarter of its cells. Hydrogen flows through the hypodermic needles, and air passes through the remaining cells. At the exit of the burner, the gas streams mix and create a flat flame just above the burner surface. The inner Hastalloy honeycomb is surrounded by a 6 mm wide nitrogen coflow, which is separated from the main flow by a 0.73 mm wall. The nitrogen coflow not only improves flame stability in the center of the burner, but also reduces the effects of room air on the flame itself.

Three MKS Alta-Series mass flow controllers were used to set the gas flow rates. Flow rates for the burner were scaled to match the conditions used by Hancock et al. [7] to create a large region of uniform flame from 1.5 to 5.0 cm above the burner surface. Hancock utilized \(N_2\) CARS thermometry to show that temperatures remained within 1% of the adiabatic flame temperature over this axial range [7]. The high flow rates allowed the flame to stabilize at a sufficient height from the burner surface to ensure that heat losses to the surface were minimized during combustion. For the present experiments, the airflow rate was maintained at 40 standard liters per minute (SLPM), while the hydrogen flow rate was changed from \(~6\ to 24\) SLPM to produce equivalence ratios ranging from 0.4 to 1.4. Nitrogen flow was maintained at \(~20\) SLPM to provide flame stability and isolation from the surrounding air. The accuracy of the mass flow controllers was \(\pm 1\%\) of the set point, with a repeatability of 0.2% of the reading. All flows were calibrated using a Bios International Definer 220 H flow meter.

Excitation of the OH transitions was performed using the Nd:YAG-pumped dye-laser and frequency-doubling system shown in Fig. 2. The 1064 nm output of a Spectra-Physics QuantaRay Nd:YAG laser was passed through an external second-harmonic generator. The resulting 532 nm beam then pumped a Continuum ND6000 dye laser with a dual grating, allowing a minimum line width of about 0.06 cm\(^{-1}\) at 560 nm. Finally, the generated dye-laser beam was passed through another doubling crystal that was located within a UVT-1 automatic tracking unit, through which both the residual dye beam and the generated UV beam exited and were separated via a Pellin Broca prism located within the exit of the autotracking unit.

The residual dye-laser beam was used to track the wavelength. The collimated beam travelled through a beam splitter, where 1% of the beam entered a 3.5 optical density neutral-density filter before being
focused into a single-mode optical fiber. The signal was transmitted through the optical fiber to a High Finesse WS-7 wavemeter, which provided measurement of the wavelength with an absolute accuracy of about 60 MHz for the wavelengths used. The wavemeter also measured the line width of the dye-laser beam.

The UV beam was passed across the top of the burner using a periscope and a set of sheet-generating UV fused-silica lenses. An \( f = -300 \) mm cylindrical lens coupled to an \( f = 1000 \) mm spherical lens created a focused sheet \( \sim 2 \) cm in height above the burner, where the beam excited the OH transitions. An image-intensified Princeton Instruments PI-MAX2 ICCD camera detected the fluorescence from the excited OH. Triggered by a 10 Hz pulse that was generated by an external timing unit, a Princeton Instruments programmable timing generator (PTG) controlled the intensifier pulse and delay time. The intensifier pulse width was set to 40 ns, which aided the elimination of chemiluminescence from the flame. The fluorescence passed through a CG-295 Schott glass filter to eliminate laser scatter before entering a 65 mm UV Nikon Lens attached to the PI-MAX camera.

### 3. Simulations

The equilibrium flame temperature and relevant species concentrations at each equivalence ratio were determined using the STANJAN [20] thermodynamic equilibrium software, which employs reference data from the JANAF thermochemical tables [21]. Equilibrium concentrations aided the determination of the quenching corrections of the LIF signal (to be described in Section 4). Spectral simulations were performed using LIFBASE 2.0.60 [22]. LIFBASE software was used to identify line locations, relative intensities, and line widths at different temperatures. The central wavelengths of various OH transitions used for the measurements are provided in Table 1.

#### A. Line-Peak-Ratio Simulations

Simulations of the spectral lines with temperatures are a critical part of thermometry. Line ratios were determined by first extracting the line intensities of each rovibronic line at various temperatures. The contribution to the line shape from collisional broadening was based on measurements performed by Rea et al. [23]. The simulated fluorescence intensities of the desired line pairs were then compared with the experimentally measured ratio. For the present study, line pairs were selected based on previous research that was discussed in Section 1, and additional line pairs were added to examine the importance of selecting spectrally isolated transitions.

Simulated line ratios of various OH transitions for temperatures ranging from 1000 K to 3000 K were determined using LIFBASE. A curve fit to the data for each line pair relates the line intensity ratio, \( r \), to temperature, \( T \). The quality of the curve fit is important because the expression determined must accurately represent the simulated data to ensure proper extraction of temperature from the experimental data. A fifth-degree polynomial fit was performed on all of the correlated line ratios. These curve fits were then used to determine the temperature from the experimentally determined line-peak ratio, \( r \). Relationships of the line ratio to temperature have been provided by other researchers, such as Seitzman et al. [14] and Devillers et al. [24], which provide information on two-line line-peak thermometry optimization. As according to the relationships in the literature, the \( Q_2(8)/Q_1(9) \) line ratio exhibits almost no sensitivity to temperature as determined by a very small energy difference between the ground states.

#### B. Line-Area-Ratio Simulations

Simulations for the line-scanning thermometry involved the creation of Voigt profiles for each spectral line. Contributions of collisional and Doppler broadening to the line width were calculated using LIFBASE 2.0 [22]. The Gaussian width of the instrument function was added in quadrature to the Doppler width. Areas of two different spectral lines are required for the development of a relationship

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**Table 1. Line-Center Locations Determined from LIFBASE 2.0 [22]**

<table>
<thead>
<tr>
<th>Line</th>
<th>Vacuum Wavelength (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_2(8) )</td>
<td>281.725</td>
</tr>
<tr>
<td>( R_2(5) )</td>
<td>281.738</td>
</tr>
<tr>
<td>( R_2(13) )</td>
<td>282.639</td>
</tr>
<tr>
<td>( P_1(2) )</td>
<td>282.663</td>
</tr>
<tr>
<td>( Q_1(5) )</td>
<td>282.750</td>
</tr>
<tr>
<td>( Q_1(9) )</td>
<td>284.005</td>
</tr>
<tr>
<td>( Q_2(8) )</td>
<td>284.009</td>
</tr>
<tr>
<td>( P_1(5) )</td>
<td>284.028</td>
</tr>
<tr>
<td>( P_1(7) )</td>
<td>285.088</td>
</tr>
<tr>
<td>( Q_2(11) )</td>
<td>285.157</td>
</tr>
<tr>
<td>( Q_1(14) )</td>
<td>286.456</td>
</tr>
</tbody>
</table>
between gas temperature and the line-area ratio. The area, $S$, of each individual line was calculated using the integral in Eq. (1):

$$ S = \int_{\lambda_0 - \Delta\lambda_i}^{\lambda_0 + \Delta\lambda_i} F(v) dv, $$

where $\lambda_0$ is the central line position and $\Delta\lambda_i$ provides a range for the integration. $F(v)$ is the simulated fluorescence line shape that is determined by a Voigt profile for each line. Figure 3 illustrates how this integral can be applied to the $Q_1(5)$ transition, with the integration range displayed here only serving as a graphical representation, the actual integration width used is described in Section 5. Simulated line-area ratios were determined relating temperature as a function of the area ratio of two spectral lines and the integration range, $\Delta\lambda_i$. The integration widths for both transitions used in determining the area ratio were set to the same value; i.e., $\Delta\lambda_i$ is the same for both transitions. Application of Eq. (1) to experimental data requires the use of a curve fit of the data, from which integration of the resultant fit will result in the line area. Discussion of the effects of the integration range on the experimentally determined temperature will follow in Section 7.

4. Quenching and Power Corrections

The fluorescence signal from LIF measurements performed within a linear regime is represented by Eq. (2) [1]. All measurements of fluorescence were conducted within the linear regime, which was confirmed via laser-energy scans of each transition used for measurements:

$$ F = \frac{h\nu \Omega}{c} \frac{VN B_{12}}{4\pi} \frac{A}{A + Q}. $$

The terms in Eq. (2) that are constant for a given experimental setup cancel when a ratio of two fluorescence signals is calculated. These constants are $h$ (Planck’s constant), $\nu$ (laser frequency), $\Omega$ (solid collection angle), and $c$ (speed of light). The fluorescence signal reduces to Eq. (3), which shows that the fluorescence is linearly proportional to the spectral beam intensity ($I_0$), the population ($N$), and the LIF efficiency, $A/(A + Q)$:

$$ F \propto VN B_{12} \frac{A}{A + Q}. $$

Fluorescence measurements for determining the species concentration of OH must be corrected for changes in laser power and LIF efficiency. LIF-efficiency values are dependent on quenching, which is a nonradiative exchange of energy from the excited OH species to surrounding species. Quenching corrections require a LIF-efficiency value that is determined using the following method. The value of $A$, the spontaneous-emission coefficient, for OH is acquired from LIFBASE for the appropriate lines. The quenching rate, $Q$, must be known for each equivalence ratio used in the measurements. The predictions from equilibrium combustion calculations provide the equilibrium concentrations of the surrounding species. If the concentration of each species, the temperature, and the pressure are known, Eq. (4) yields the quenching rate:

$$ Q = \sum_i \left( \frac{P}{kT} \right)(\tau_i), $$

where the quenching-rate coefficient, $\tau_i$, for each species, $i$, is provided by Tamura et al. [25]. Corrections for quenching to all LIF measurements were performed by dividing the LIF signal by the appropriate LIF efficiency. As temperature is required to determine the quenching rate, an iterative procedure was used with the equilibrium temperature as the initial guess. Laser-power corrections were determined by direct measurement of the UV laser beam power. Because of the large near-adiabatic region created above the Hencken burner that was used for the measurements, fluorescence signals were extracted within a uniform-intensity region located at the peak of the planar LIF (PLIF) imaging field. Figure 4 shows the PLIF signal and its location relative to the heights where Hancock found near-adiabatic conditions, between 1.5 and 5 cm [7] (represented by the distance between the dashed lines in Figs. 4). Within the peak region of laser intensity where the fluorescence signal was collected, an RMS temperature error of only 3% was found, based on beam intensity nonuniformities.
5. Experimental Results

A two-line line-peak-ratio thermometry technique was used for temperature measurements with the various line pairs described in Section 1; the pair with the least difference to calculated temperature values will be used for line-area-ratio thermometry. Each measurement consisted of acquiring 50 individual images, and each image resulted from 10 on-chip accumulations of pulses. Following data collection, the 50 images were averaged; and a background image, acquired under the same conditions with the flame extinguished, was subtracted. Intensities were extracted from a uniform-intensity region, depending on the beam profile, within the PLIF image. The area of the uniform region varied from 6 to 10 mm in the horizontal direction and from 1 to 4 mm in the vertical direction. Figure 5 shows an example of a horizontal slice through the image in Fig. 4, taken from a region of uniformity in the vertical direction. Statistics on the region yielded a mean value that was used as the associated line intensity, along with an RMS value of the noise associated with each value. A significant source of error was the shot-to-shot variation of intensity among the 50 acquired images. Average values within the same integration window were extracted from each image. The fluctuation of these average values was \( \sim 4\% \). This experimental error aided determination of the temperature variation associated with a Boltzmann-fraction change of 4% for each line pair used. Error bars of each measurement include errors associated with this signal-to-noise and shot-to-shot laser-power fluctuations.

6. Line-Peak Temperature Measurements

Application of the curve fits determined from simulations to the acquired LIF intensities yielded temperature profiles as a function of equivalence ratio. With corrections of laser power and LIF efficiency, a minimum temperature error of \( \sim 6\% \) was present using the \( Q_1(14)/Q_1(5) \) line pair. These temperature measurements are subject to errors that result from absorption differences between the lines, as well as from spectral-overlap differences caused by the inability to tune the dye laser to the precise wavelength of a line peak.

A. Absorption Corrections

Absorption of the laser beam as it passes through the flame affects the laser intensity that is present where fluorescence has been extracted. The laser intensity across the flame varies according to the Beer–Lambert law and is a function of the local OH concentration and the absorption coefficient of the transition of interest. Variations of the absorption between different transition lines due to different absorption coefficients can result in errors when two lines are used in a ratio-based measurement. To determine the attenuation of the laser beam at each spectral line, an OH-concentration profile was required. Absorption for each line was estimated using a normalized profile of the LIF intensity across the flame as a shape function for the OH concentration. Normalization was performed by dividing the LIF intensity across the flame by the average of intensities ranging from \(-1.0\) to \(1.0\) cm. This normalized profile, Fig. 6, provided a relative profile of OH, but still did not provide a quantitative measure of OH concentration. To extract an OH-concentration profile, an equilibrium OH-concentration value from STANJAN was multiplied by the normalized shape function.

The laser intensity across each flame front was then determined by applying the Beer–Lambert law to the OH concentration. Absorption coefficients

Fig. 4. (Color online) PLIF signal above the exit of the Hencken burner.

Fig. 5. Horizontal slice of PLIF intensity for determination of uniform interrogation region.
were extracted from LIFBASE 2.0 [22]. An absorption correction factor was determined by averaging the intensity ratio across the region of extracted fluorescence, which ranges from $-0.5$ to $0.5$ cm in Fig. 6.

B. Spectral-Overlap Corrections
As the dye laser is tuned to various absorption transitions, the spectral overlap of the laser profile can vary, depending on its position with respect to the absorption peak, as well as to the possible effects of neighboring transitions. The spectral overlap, $g_o(v)$, of each measurement was determined using Eq. (5):

$$g_o(v) = \int_{-\infty}^{\infty} g_l(v_l)g_a(v_a)dv.$$

The laser line shape is given by $g_l(v_1)$, for which a Gaussian line shape was used with a central wavelength, $v_1$, and a line width of about 0.09 cm$^{-1}$, as determined by the use of the wavemeter described in Section 2. The absorption-line shape, $g_a(v)$, included both Doppler and collisional-broadening terms, along with the contribution of neighboring transition lines. The contribution of these lines to spectral overlap was determined by incorporating the sum of multiple absorption line shapes, each centered at the respective line position and normalized by the relative peak intensities found using LIFBASE [22]. Figure 7 contains a sample of each line shape described and shows the $Q_1(5)$ line, with the $Q_{21}(5)$ neighbor present in the absorption-line profile. Correction of the fluorescence measurement required the multiplication of each spectral-overlap value by the measured fluorescence intensity.

The measured temperature after incorporation of corrections for power, quenching, absorption, and spectral overlap, is presented in Fig. 8. Temperature values were compared to STANJAN equilibrium-temperature values over an equivalence-ratio range of 0.4–1.4. The $Q_1(14)/Q_1(5)$ line-ratio measurements yielded the highest accuracy relative to the calculated equilibrium values, with an average difference of 3.6% over the equivalence-ratio range. The $R_{2}(13)/P_{1}(2)$ line pair was also employed, with...
accuracies of about 4.1% being achieved. All of the line ratios exhibited improved results when stoichiometric conditions were approached, especially the $R_2(8)/Q_2(11)$ and $P_1(7)/Q_2(11)$ line pairs, which were used by Seitzman et al. [14]. Other line pairs employing $Q_2(11)$ (not presented in Fig. 8) resulted in higher temperature differences, as observed by Seitzman et al.

The temperature difference from equilibrium values at lower equivalence ratios, ranging from 0.4 to 0.8, were higher than those for equivalence ratios ranging from 0.9 to 1.4; for example, within these ranges, the differences for the $R_2(8)/Q_2(11)$ pair were $\sim 16\%$ and $8\%$, respectively. The considerably lower signal from the Boltzmann-sensitive line at these low-equivalence ratios is a source of error as noise becomes more significant. RMS noise of the extracted $Q_2(11)$ fluorescence intensity reaches a value of almost $10\%$ at an equivalence ratio of 0.4, as compared to a value of about $4\%$ at an equivalence ratio of 1.0. The absorption correction of each line pair is also a source of error at various equivalence ratios. All absorption corrections were performed under the assumption that the OH concentration at the location of fluorescence was equal to the equilibrium OH concentration. However, in a region above the Hencken burner, superequilibrium, or values greater than equilibrium, can exist [18]. Simulations performed by Kulatilaka showed that superequilibrium concentrations grew as equivalence ratios departed from 1 [18].

To study the sensitivity of each line pair to the superequilibrium concentration of OH, absorption corrections based on superequilibrium concentration rather than equilibrium concentration of OH were used. Figure 9 shows the temperature gradient associated with each line pair at various superequilibrium values used for absorption at an equivalence ratio of 1.0 (in Fig. 9, SE is defined as the ratio of actual OH concentration to equilibrium concentration). The two line pairs, $Q_1(14)/Q_1(5)$ and $R_2(13)/P_1(2)$, with the lowest sensitivity to OH concentration were also the two pairs that yielded the highest overall temperature accuracy. Under stoichiometric conditions, changing the superequilibrium values from 1 to 2 when using the $R_2(8)/Q_2(11)$ pair would result in an almost 400 K temperature change. Equilibrium concentration under stoichiometric conditions is orders of magnitude larger than that at an equivalence ratio of 0.4. Doubling the superequilibrium value creates a much larger change under lean conditions. For this reason, the equilibrium assumption can cause greater deviations in measured temperatures at lower equivalence ratios.

As stated in Section 1, a good rule to follow regarding line selection is to use isolated lines for line-ratio measurements. To illustrate how temperature measurements can vary when the lines are not isolated, three line combinations are used. The first pair, $Q_2(8)/Q_1(9)$, represents a line pair with the lines being neighbors in close proximity, about $3.4\,$pm, which is similar to the line width of the dye-laser beam. The second pair has only one line with a neighbor, the $P_1(5)/Q_1(9)$ line pair. It is the $Q_1(9)$ line that is affected by its neighbor, the $Q_2(8)$ line. The measured temperatures based on these two line pairs will be compared to that found using the third line pair, $Q_1(14)/Q_2(15)$, located at 286.456 nm and 282.75 nm, respectively. It will be shown that this proximity is a possible cause of large deviations in measured temperatures from calculated equilibrium temperatures.

![Figure 9](image9.png)

**Fig. 9.** Temperature sensitivity as a function of superequilibrium OH concentration for an equivalence ratio of 1.0.

![Figure 10](image10.png)

**Fig. 10.** Extracted temperatures from closely spaced and isolated spectral lines of OH.
Temperature values for these three line pairs are presented in Fig. 10. As expected, the $Q_1(9)/Q_2(8)$ pair, with line proximity similar to the laser line width, yielded temperatures with errors larger than 300%. The $P_1(5)/Q_1(9)$ pair provided more reasonable temperatures; however, the errors were still large compared to those of the $Q_1(14)/Q_1(5)$ pair. Errors can be due to the effects of the $Q_2(8)$ line on the $Q_1(9)$ line used for the intensity ratio. These large errors support the rule that Seitzman et al. established concerning selection of lines [14]. It is important to note that the $Q_1(5)$ line also has a neighboring transition, the $Q_{21}(5)$ line, which is located about 10 nm from the $Q_1(5)$ line and is about three times the laser line width. The $Q_{21}(5)$ line, however, has the same rotational number as the $Q_1(5)$ line, leading to a lower effect on acquired temperatures. When considering the Boltzmann-fraction population effects of each of the three line pairs on temperature, the $Q_1(14)/Q_1(5)$ pair yielded the lowest temperature difference based on errors in the Boltzmann fractions, followed by $P_1(5)/Q_1(9)$ and $Q_1(9)/Q_2(8)$ pairs, in the same order as the relative accuracies of the equilibrium-temperature values.

When corrections for absorption and spectral overlap were not performed, the $Q_{1}(14)/Q_{1}(5)$ line pair yielded a measured temperature difference of about 9% when compared to calculated values, averaged over the entire equivalence-ratio range employed. When the LIF signal was corrected for the absorption only, the difference decreased to $\sim 8.6\%$. The deviation decreased to $\sim 4.5\%$ when the signal was corrected for the spectral-overlap function only. Since the latter correction had a greater effect on the resultant error of the $Q_{1}(14)/Q_{1}(5)$ line pair, it should be implemented, if possible. If the highest possible accuracy to calculated temperatures is required, however, both corrections should be performed; for this line pair, achievement of an error as small as 3.6% is possible.

7. Line-Scanning Measurements

The line-area-ratio study was performed using the $Q_1(5)$ and $Q_1(14)$ transitions. Temperature data using this line pair was extracted by using the line-area and temperature relationship described in Subsection 3.B. Line shapes were determined by scanning the wavelength of the laser across the transition line using a 1–2 pm step size. A background image was acquired after each step and subtracted from the raw image. A single background subtraction following a complete scan is one possibility to speed acquisition; however for this study, the effects of background variations were minimized by performing this subtraction following each data point. An LIF intensity value was obtained from each background-subtracted image using the method described in Section 5 for two-line line-peak-ratio thermometry. Laser-energy normalization of each intensity measurement involved the division of LIF intensity by the laser power measured at each step.

Quenching corrections for each scan were constant, as the equivalence ratio during each scan did not change.

To determine the line shape of each line used for the line-area-ratio method, a Levenberg–Marquardt fitting algorithm was implemented, allowing for Voigt fitting of the acquired data. The line fit for the $Q_{1}(14)$ transition involved a single Voigt fit; however, due to the presence of a neighboring line, the $Q_{21}(5)$ as stated in Subsection 6.B, fitting of the $Q_{1}(5)$ for extraction of an area required using the sum of two Voigt profiles, each being centered at the respective transition wavelength. The fitting of the $Q_{1}(14)$ line was performed using a single Voigt function centered at 286.456 nm. A sample fit of the data acquired for the $Q_{1}(5)$ transition, which includes contribution from the $Q_{21}(5)$, is displayed in Fig. 11, while a sample fit for the $Q_{1}(14)$ is shown in Fig. 12.

Area ratios of the data were determined using the technique described previously in the line-scanning-simulation section (Subsection 3.B) of this paper. Two line areas were determined using Eq. (1), each using the same integration range. The ratio of the two areas was then used to extract a temperature based on simulated area-ratio relationships to temperature. It is important to understand how the selection of an integration range may affect the value of the extracted temperature; this measured temperature dependence on integration range, $\Delta \lambda$, is shown in Fig. 13. As the integration range is varied from zero to about 20 pm, the measured temperature asymptotically approaches a steady value. The integration range needed to acquire an accurate temperature is dependent on the overall line width of the transitions employed. As a general guideline, the integration width should be selected such that the change in the area ratio versus the integration width

![Image](image-url)
used for the area ratio must approach zero, while the change in each transition area versus the integration width must also be zero, or approach a constant value. Figure 14 presents an example of the relationship between the area and the integration width used for individual transitions, $S_1$ and $S_2$, for the area of the $Q_1(14)$ and $Q_1(5)$ transitions, respectively, using Eq. (1). Because of the neighboring transition of the $Q_1(5)$ transition, the area does not approach a steady-state value as soon as that of the $Q_1(14)$. This difference also contributes to the shape of the $S_1/S_2$ curve in the figure, which is similar in shape to the temperature curves seen in Fig. 13. The shape of these curves will be different depending on the transitions selected for the line-area-ratio method, with isolated transitions yielding area trends similar to that of the $Q_1(14)$ area, $S_1$, and transitions which are not isolated yielding curves that will vary depending on the strength and location of neighbors. When attention is focused on the $S_1/S_2$ curve, it can be seen that, as the integration width becomes large, the change in this ratio becomes zero. There is, however, also a region around 5 pm that has a zero slope, suggesting that finding an integration width in which the change in the area ratio versus the integration width is zero may not be enough to set a proper width. This leads us to the final curve in Fig. 14, $S_1 \cdot S_2$, or the product of the line areas. The product of the areas will help determine if both line areas have reached a steady-state value at a given integration width; when the product has reached a steady-state value, Eq. (6), then a suitable width has been selected:

$$\partial (S_1 \cdot S_2)/\partial (\Delta \lambda_i) = 0.$$  \hfill (6)

If a neighboring transition is present, this integration width increases as $\partial S_i/\partial (\Delta \lambda_i)$ does not approach zero, until the width is further increased to include the neighbor. All measurements taken must span a range that is sufficient to capture the line shape of each transition that falls within the integration range needed for line-area-ratio measurements. For simplicity, it may be beneficial to use two completely isolated lines for the line-area-ratio technique to reduce the required experimental scan range. This will not only allow for the use of one single Voigt fit for each transition, but might also allow for the use of a simpler representation of each fit in which, instead

Fig. 12. Excitation-scan data and fit for the $Q_1(14)$ line at $\phi = 1.0$.

Fig. 13. Measured temperature as a function of the integration width of the peaks.

Fig. 14. Measured line areas $S_1$ and $S_2$ along with their ratio and product as a function of the integration width.
of the area, relationships between fit parameters of each transition pair to temperature could be used [26]. The temperatures obtained using the line-area-ratio method and the line-peak-ratio technique described in Section 3 are plotted in Fig. 15. The temperatures obtained using the former method yielded a higher accuracy (2.4% average difference) than those obtained using the latter (6% average temperature difference) across the full range of equivalence ratios. The difference between these results was greater at lower equivalence ratios. At an equivalence ratio of 0.4, for example, the line-area-ratio measurements reduced the difference in the line-peak ratio from 19% to 6%. The attributed gain in accuracy may be due to the effects of a low signal-to-noise ratio at a low equivalence ratio, combined with variations in collisional parameters. Reduced differences between the experimental temperatures and the calculated values could be contributed to the required data fitting of the line-area-ratio method, which reduces the effects of low signal-to-noise at reduced equivalence ratios. Please note that, at low-equivalence ratios, the upper limit of the error bars are always closer to the theoretical value, which is not due to systematic error in the experiment but may be just a coincidence.

8. Conclusions
Temperature measurements were performed using both a two-line line-peak-ratio method and a two-line line-area-ratio technique and the results compared. The line-area-ratio method was found to provide temperatures with an average temperature difference, to calculated values, of ~2.4% over a range of equivalence ratios from 0.4 to 1.3. Integration-range effects on the measured temperature were discussed, and it was shown that the integration range must be such that any further increase in the integration range will not change the measured areas of each transition, represented by Eq. (6). The line-peak technique produced greater differences at lower equivalence ratios because of the reduction in signal-to-noise ratio at lower temperatures. At equivalence ratios around one, however, the line-peak technique produced results sufficiently similar to those of the line-area technique that little justification could be found for the added acquisition time and data processing required to extract temperatures from the line-area-ratio method. Use of a high-speed camera along with a UV intensifier, as well as streamlining the data acquisition processes for the line-area-ratio method, such as simultaneous scanning and acquisition, will significantly reduce the data acquisition time and make this approach suitable for investigating turbulent reacting flows. If measurements for an expanded range of equivalence ratios are required, it is suggested that the line-area-ratio method be implemented, if possible, due to its improved experimental accuracy. For line-peak-ratio measurements, the \( Q_{1(14)}/Q_{1(5)} \) line ratio provided the most accurate results after the signal was corrected for spectral overlap and laser absorption. This line ratio also exhibited the least sensitivity to errors in absorption concentration, which indicates that, when absorption corrections are performed, an estimated value for OH concentration is sufficient to provide accurate results. The ratio of LIF signals for the \( Q_{1(14)}/Q_{1(5)} \) line pair without spectral-overlap and laser-absorption corrections yielded a temperature error of ~9%. However, the spectral-overlap correction improved the temperature measurement accuracy to within 4.5% over a 0.4–1.4 equivalence-ratio range when compared to calculated equilibrium temperatures.

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