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Published in:
E1-2-111

2015

Link to publication

Citation for published version (APA):
Sprays thermometry using two-color LIF and SLIPI

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Abstract

Knowing the droplets temperature is of great importance in spray-assisted combustion devices as it directly affects the droplet evaporation rate. This information helps optimizing the fuel/air mixture formation for the improvement of combustion devices. To measure the temperature of spray droplets, optical techniques such as Rainbow-Refractometry (for point-wise) and the two-color LIF ratio thermometry (for 2D) have been preferred to thermocouples due to their non-intrusive nature. However, in optically dense sprays, these techniques are affected by unwanted impedes such as multiple scattering of light, intensity reduction along the light propagation (laser extinction) and attenuation of the signal travelling from the incident plane to the detector. The multiple light scattering contains false signals emerging from the non-illuminated portions of the sprays and leads to erroneous measurement results as well as image blur. Thanks to SLIPI (Structured Laser Illumination Planar Imaging) these issues can be efficiently addressed. In addition, by using two-color LIF (Laser Induced Fluorescence), effects of laser extinction and signal attenuation can be canceled out from the image ratio.

In this study, SLIPI is combined with the two-color band LIF ratio approach using a temperature sensitive dye. Here fluorescein is excited at 447 nm and emits a LIF signal peaking at 520 nm. This signal becomes red-shifted as the temperature increases. We demonstrate, for the first time, the application of this novel approach for measuring the local temperature of a hollow cone (HC) spray in 2D. The water temperature in the presented measurements ranges from 20°C to 90°C while the liquid injection pressure is set to 20 bars. The measurement is calibrated in-situ with a thermocouple and the results from both the conventional and SLIPI two-color LIF detection schemes are compared. It is found that the SLIPI ratio shows a significant improvement in signal sensitivity and accuracy when compared to the results of the conventional planar imaging.

Keywords: spray imaging, temperature mapping, multiple scattering, SLIPI

1. Introduction

Control and optimization of the evaporation rate of liquid droplets are desired when atomizing sprays are especially used in internal combustion (IC) engines, spray cooling and spray drying applications. For example, in IC engines, an evaporation rate of few milliseconds is required for the fuel droplets to quickly vaporize and form the adequate fuel/air mixture proportion before ignition. In spray cooling, uniform cooling is necessary because the surface heat flux varies spatially. Thus, the heat transport phenomenon in sprays is quite complex and to measure the evaporation rate precisely, droplet temperature must be known with greater accuracy. Several non-contact (optical) and contact (thermocouples) methods have been developed so far to measure temperature of liquids and sprays [1, 2]. Nevertheless, the optical techniques are preferred because they can provide point-wise as well as full-field mapping of temperature fields without disturbing the medium.

Optical methods such as Rainbow Refractometry, Raman scattering, fluorescence lifetime, laser-induced exciplex fluorescence (LIF) and Laser Induced Fluorescence (LIF) have been applied for thermometry [2] of liquids. Some less common alternative optical approaches are Morphology-dependent resonances (MDRs), thermographic phosphors, Molecular tagging thermometry, thermochromic liquid crystals (TLC), infrared thermometry [2]. The Global Rainbow refractometry deduces the temperature by correlating it with droplets refractive index. However, the presence of non-spherical droplets give large measurement uncertainties and it is rather time consuming to implement in sprays. The TLC thermometry was demonstrated by Richards et al. on evaporating cooling water droplets [3]. In this, chiral-nematic liquid crystals refract light of selected wavelengths as a function of their temperature. In LIF thermometry, the intensity ratio of exciplex and monomer extracts the temperature of the liquid. This approach requires an oxygen free environment to avoid exciplex quenching and therefore, it is impossible to implement in spray-assisted practical combustion devices.

The laser sheet based LIF thermometry approach is very attractive because it is capable of extracting a 2D map of temperature of an illuminated section with high spatial and temporal resolution. Using two-color LIF ratio method, 2D mapping of temperatures in liquids, droplets and sprays have been reported [4-10]. In order to apply it, one and/or more dyes are judiciously selected which produce a temperature sensitive spectral shift in their LIF signals when excited with an appropriate wavelength. However, the two-color (one dye) based ratio
scheme is preferred because it eliminates the issue of changes in dye concentration, laser intensity fluctuations etc. [6-8]. Lemoine and co-workers have pioneered this method for thermometry in droplets steam [6-8]. Nevertheless, impedes such as the nonlinear effect of droplet size on the LIF intensity ratio (at constant temperature, it increases significantly as droplet size reduces) has been reported while applying it in sprays [11]. It is demonstrated that the non-linear size effect and multiple scattering effects lead to a bias in the ratio. In a recent article using SLIPI-LIF/Mie [12], Mishra et al. demonstrated on an HC water spray that prior to intensity ratio, LIF and Mie images must be corrected for effects from multiple scattering (even in dilute spray conditions) for reliable measurements of SMD. By means of using SLIPI such discrepancies were efficiently removed. Thus, in this work, we have reported for the first time, a novel approach combining SLIPI and two-color (single dye) LIF ratio thermometry on an HC spray. The presented work is a further continuation of Polster et al. [13] who successfully demonstrated the technique on a transparent cuvette for water temperature ranging between 20 to 90 degrees. Here, it is applied in a spray set at 20 bars of liquid injection pressure for a temperature range of 20 to 50 degrees. The SLIPI-LIF ratio is correlated as a function of temperature. Based on this, 2D images of absolute temperature are extracted.

2. Description of the imaging techniques
2.1 Two-color LIF ratio thermometry

The intensity ratio of two-bands of a LIF signal showing spectral shift either with an increase or decrease in the temperatures of the dye-doped liquid are extracted. To achieve a quantitative measurement map of the temperature map, this ratio is calibrated with the temperature measured by a thermocouple. A correlation between the ratio and temperature is plotted. Consider that \( I_{LIF1} \) and \( I_{LIF2} \) are the LIF intensities of two bands at liquid temperature \( (T) \). The two-color LIF ratio \( (R) \) after pixel by pixel overlapping and with background correction can be expressed as:

\[
R(T) = \frac{I_{LIF1} - BG_1}{I_{LIF2} - BG_2}
\]  

(1)

Here, \( BG \) is the background image, which is recorded when the laser sheet is not exciting the dyed liquid. By combining this method with SLIPI (described in section 2.2), it is aimed that a better dynamics and accuracy in the measurement can be achieved after suppression of multiple scattering from images of two bands prior to intensity ratio.

2.2 SLIPI

Structured Laser Illumination Planar Imaging (SLIPI) is inspired from structured illumination microscopy [14]. It is developed at Lund University for imaging dense spray systems (where the majority of photons leaving the medium suffers from the multiple scattering) [15]. A sinusoidal modulation (vertically) is spatially imprinted on the conventional laser sheet using a Ronchi grating and a spatial filter. When intersecting a section of an atomizing spray (a scattering medium), the photons that scatter only once (known as singly scattered) do “remember” this signature while the photons which interact with the droplets more than once (known as multiply scattered photons) “forget” this modulation. Thus, using a post processing computation algorithm, a conventional image (comprising of both the singly and multiply scattered photons) and a SLIPI image represented only by the singly scattered photons can be extracted. If a sinusoidal pattern is impressed on a light sheet, the resultant intensity of the image \( I(x, y) \) is described as:

\[
I(x, y) = I_c(x, y) + I_s(x, y) \cdot \sin(2\pi x v + \phi)
\]  

(2)

Where, \( v \) represent the modulation frequency and \( \phi \) the spatial phase. Here, \( I_c(x, y) \) is the intensity corresponding to singly and multiply scattered photons and, \( I_s(x, y) \) represents the amplitude of the modulation from the singly scattered photons only. In order to deduce this modulation over a full image and to experimentally extract the intensity component of \( I_s(x, y) \) in Equation 2, a minimum of three intensity modulated recordings (sub-images: \( I_0, I_{120}, \) and \( I_{180} \), each with a spatial phase difference of \( 120^\circ \), are required. A SLIPI image is constructed by means of taking the root-mean square of the sub-images while a conventional image can be deduced by averaging the same.

To maintain the very high spatial resolution of the images and to remove all the identical components in subsequent recordings, a minimum of three sub-images are always needed. A detailed description of the SLIPI technique and its uses in 2D imaging of sprays can be found in [16]. A two sub-image-based SLIPI setup developed for imaging spray dynamics at the cost of spatial resolution has been recently reported [17].
3. Description of the experiments
3.1 Absorption of fluorescein

A commercially available concentrated solution of Fluorescein 27 (FL 27) (7% by weight) dye (Kingscote chemicals, USA) has been used. These are biodegradable and non-toxic according to the manufacturer. This is the main purpose of using this dye in addition to its high quantum efficiency at 447 nm. Sutton et al. has demonstrated that FL 27 has a positive temperature sensitivity (3.5 %/°C), significantly higher than Rhodamine B and Kiton Red (-1.6%°C) for an excitation wavelength of 532 nm [18]. FL 27 gives positive as well as negative temperature dependence at different excitation wavelengths [13], therefore, it is an excellent dye for LIF thermometry, giving more flexibility depending on the application. The absorption spectrum of the dye solution is recorded in a photometer (T60U, PG Instruments). The absorption given in Fig. 1(a) shows maximum at 485 nm as well as additional peaks at 323 nm, 283 nm and 236 nm. The molecular structure of the dye is shown in figure 1(b).

![Absorption spectra of fluorescein dye](image1.png)

**Figure 1** (a) Absorption spectrum of fluorescein dye (dissolved in water) is given. (b) The molecular structure of fluorescein.

3.2 Response of the Fluorescein emission

The spectral temperature-dependent properties of the dye are investigated at an excitation wavelength of \( \lambda = 447 \) nm and at temperatures ranging from 20 °C to 90 °C. It is recorded using a spectrometer (AvaSpec-USB2 version 7.6.1) and a thermocouple [13]. The emission spectrum as a function of non-normalized and normalized intensity against wavelength for 447 nm is shown in figures 2(a) and 2(b), respectively.

![Emission spectra of fluorescein dye](image2.png)

**Figure 2** The Non-normalized and normalized LIF emission spectra of the dye at 447 nm in a temperature range from 20-90 degrees is shown in figures 2(a) and 2(b), respectively. A change in LIF emission intensity in (a) and spectral shift (b) with variation in temperature is apparent.

In figure 2(a), the maximum fluorescence intensity of 22000 photon counts decreases by 27.3 % with an increase in temperature from 21 to 90 degrees. When normalized, the spectrum in figure 2(b) indicates a red shift of the intensity maximum from 523 nm to 530 nm, which is especially distinct on its right side. This is due to the fact that the spectral emission broadens and also slightly changes its shape with increasing temperature.

3.3 Temperature sensitivity for different spectral band ratios

In order to achieve optimized temperature sensitivity, selection of a ratio of the two spectral bands is plotted as a function of temperature, where the slope of the graph indicates the sensitivity of the measurement. This sen-
sitivity depends on certain parameters, e.g. the excitation wavelength and the temperature-dependent properties of the tracer. Therefore, the spectral band ratios are selected on the following basis:

(i) Two-color LIF ratio using large spectral bands: to determine the highest signal intensity response, i.e. to optimize the amount of light intensity. This results in a small spectral band to the left of the intensity maximum (left band) and a large spectral band to its right (right band). Representing the exact central wavelengths ($\lambda_L, \lambda_R$) and full-width at half maxima (FWHM) as $\Delta_L, \Delta_R$ for left bands and right bands, respectively.

(ii) Two-color ratio using small spectral bands: the determination of the best possible temperature sensitivity. An optimum sensitivity is derived from varying the width and position of the spectral bands and by calculating the ratio of those changed bands for each temperature in a range of 20 °C to 90 °C.

Looking at the different combinations of two-color LIF ratios for maximum signal intensity and temperature sensitivity optimization from [13], optical filters for experiments at 447 nm are selected. In table 1, the spectral ranges (used for the ratio) represented by groups (A, B, C) are shown.

Table 1: Spectral ranges for the calculation of the ratio for excitation wavelength

<table>
<thead>
<tr>
<th>Excitation (nm)</th>
<th>Left band (nm)</th>
<th>Right band (nm)</th>
<th>Condition</th>
<th>Selection of appropriate band</th>
</tr>
</thead>
<tbody>
<tr>
<td>447</td>
<td>$\lambda_L = 515$; $\Delta_L = 20$</td>
<td>$\lambda_R = 575$; $\Delta_R = 80$</td>
<td>A</td>
<td>Maximum Intensity (large spectral bands)</td>
</tr>
<tr>
<td></td>
<td>$\lambda_L = 507.5$; $\Delta_L = 5$</td>
<td>$\lambda_R = 612.5$; $\Delta_R = 5$</td>
<td>B</td>
<td>Maximum Sensitivity (small spectral bands)</td>
</tr>
<tr>
<td></td>
<td>$\lambda_L = 510$; $\Delta_L = 20$</td>
<td>$\lambda_R = 592$; $\Delta_R = 43$</td>
<td>C</td>
<td>Bands selected for experiments</td>
</tr>
</tbody>
</table>

Figure 3 Normalized ratio against temperature for different different band ratios at 447 nm excitation; actual values (marker) and second-order polynomial (dashed lines).

Here, A represents the normalized ratio against temperature for two bands selected for maximum intensity. Similarly, B for maximum temperature sensitivity. C represents the optimized bands selected for experiments that is a trade-off between both the high intensity and temperature sensitivity.

Using table 1, a plot of normalized ratio against temperature for the excitation wavelength is extracted using MATLAB and is shown in figure 3. For 447 nm, the maximum intensity can be achieved if the spectral band ratio (A) is extracted from left band ($\lambda_L = 515$ nm; $\Delta_L = 20$ nm) and right band ($\lambda_R = 575$ nm; $\Delta_R = 80$ nm). While
for maximum sensitivity (in B), the left band ($\lambda_1 = 507.5$ nm; $\Delta_1 = 5$ nm) and right band ($\lambda_2 = 612.5$ nm; $\Delta_2 = 5$ nm) is selected. However, in B, the FWHM bands cover only 5 nm which is not practicable for actual measurements because filters with such specification are hardly found. Hence, reasonable filters which resemble the optimized bands for the chosen excitation wavelength have to be selected. Therefore, ratio C extracted from left band ($\lambda_1 = 510$ nm; $\Delta_1 = 20$ nm) and right band ($\lambda_2 = 592$ nm; $\Delta_2 = 43$ nm) for 447 nm (see in figure 3) has been found optimum. These two fluorescence band-pass filters (Edmund Optics) ensures a transmission $>$ 93 % and a blocking at an optical density of $>$ 6.

### 3.4 SLIPI-two color LIF ratio

The 447 nm (a collimated continuous wave diode laser) incident light sheet is created from the SLIPI setup (see black dashed line in Fig. 4) that traverses a hollow-cone spray at the centre. It has dimensions: 40 mm height, 0.5 mm width and fixed at 1.5 cm below the nozzle exit. The maximum output power of the laser is set at 4000 mW. However, only 1% of it reaches at the incident plane and rest is lost by the optics and spatial filter.

To realize the two-color LIF ratio scheme, two EM-CCD cameras (denoted as LIF1 and LIF2 are kept perpendicular to each other) of the same characteristics are positioned at 90 degrees to the incident plane. The f-number ($f_\#$) of the focusing objectives is kept at 2.8. In order to record the two bands of LIF signal simultaneously, a beam splitter is used prior to using two high performance optical filters (F1 and F2) chosen for the experiments. The field-of-view of two cameras are accurately adjusted by imaging a test chart kept just at the incident plane. A perfect pixel by pixel overlap is ensured (by using a warping scheme [16]) while ratioing the two LIF intensities for thermometry. Fluorescein dye is thoroughly mixed with the tap water (1:8000 in ratio) in a tank. The dyed water is heated (using a thermocoil) before spraying it through a pressure swirl nozzle (Lechler, ordering no. 216.324) of orifice diameter 1 mm. A steady state hollow cone spray is set at a liquid injection pressure of 20 bars with a liquid flow rate of 1.26 litres/min. When the incident light penetrates the spray droplets, a broadband LIF signal peaking at 520 nm is generated. Two high performance optical filters (see specifications in section 3.4) have been used to divide a full LIF spectrum into two bands. Consequently, two SLIPI-LIF images are simultaneously recorded on the two cameras (Andor, Luca R). These 14-bit electron multiplying-CCDs provide images of 1004×1002 pixels. All the images are recorded with an exposure time of 0.88 seconds and averaged over 10 images. A thermocouple of 130 mm length and 1 mm tip diameter (K type, Pentronic AB) is kept on a 2D stage in order to insert it into the light sheet and to ensure its rapid removal after checking the temperature of droplets at that location.

**Figure 4** Experimental setup of SLIPI-two color LIF ratio thermometry. The black dashed line shows the SLIPI setup while the dashed box (red) on the top right is an illustration of two-color LIF ratio thermometry principle.
This ensures that the thermocouple tip always measures the temperature at the same position. To construct a correlation plot between temperature and LIF2/LIF1 ratio, SLIPI-LIF images are recorded for temperatures ranging between 20 °C to 50 °C with a step difference of 5 degrees.

4. Results

4.1 Non-calibrated images

The two-color LIF ratio (extracted from LIF2/LIF1) images from conventional and SLIPI schemes at reference temperatures ranging between 20 to 50 degrees are shown in figure 5(a-d) and 5(e-h), respectively. Prior to ratio, the images of the two bands are first background subtracted and then a threshold equal to 0.001 times to the intensity maxima is fixed. In order to avoid numerical errors while ratioing the data, any pixel value below this threshold is disregarded. For a fair comparison of the deduced ratio of both the conventional and SLIPI, same image processing routine is kept.

From the conventional detection (CONV) (see fig 5(a)), it is apparent that there is a false LIF ratio signal in the non-illuminated spray regions. Also, a distinct signal is emerging from the hollow-region of the spray which is unexpected due to the presence of very few and/or very small droplets. These discrepancies are generated in CONV images of sprays by effects from multiple light scattering, which hides the true value of the ratio and leads to image blur. However, in SLIPI, these effects are removed by suppressing the intensity of multiply scattered photons prior to the calculation of the ratio. The two-color LIF ratio both for the CONV and SLIPI changes with the change in liquid temperature. Also, spray symmetry can be observed in both the cases, hence, by intensity division the effects of laser extinction and signal attenuation are eliminated. Nevertheless, SLIPI shows more distinct gradient of the temperature in comparison to CONV.

![Figure 5 Averaged non-calibrated CONV-two color LIF and SLIPI-two color LIF ratio is shown in figures 5(a-d) and 5(e-h), respectively for reference temperatures 20°C, 30°C, 40°C and 50°C. In CONV-LIF ratio, images suffer from effects from multiple scattering while in SLIPI-LIF ratio such impedes have been efficiently suppressed.

4.2 Calibration

Only the SLIPI-two-color LIF ratio is considered for the calibration, mainly because of non-reliable LIF ratio extracted by CONV. In order to calibrate the SLIPI-LIF ratio, the thermocouple is inserted at a point (X = -1 cm, Y = 2 cm) given in figure 5(e). It is calibrated from 20°C to 50°C with a step difference of 5 degrees. The temperature (measured by thermocouple) is plotted against an averaged intensity ratio (20×20 matrix). The calibration showing the relationship between the SLIPI-LIF ratio and temperature is shown in figure 6. The calibration fit (a third order polynomial) is further used for extracting the absolute temperature.
Figure 6 A mean of non-calibrated SLIPI-two color LIF ratio is plotted against the droplets temperatures measured by the thermocouple. This calibration curve is used to extract the absolute 2D mapping of droplet temperature.

4.3 Absolute temperature mapping

The two-dimensional absolute temperature mapping images of spray droplets recorded at reference temperatures $T_1 = 20^\circ C$, $T_2 = 30^\circ C$, $T_3 = 40^\circ C$, and $T_4 = 50^\circ C$ are shown in figures 7(a-d), respectively.

Figure 7 The two-dimensional map of droplets temperature of a hollow-cone spray extracted from calibrated SLIPI-two-color LIF ratio is given in figures 7(a-d) for reference temperatures $T_1$, $T_2$, $T_3$ and $T_4$, respectively. Note that all the images shown here are for the liquid injection pressure set at 20 bars.
It can be seen in the figures 7(a-d), that the calibrated SLIPI-two-color ratio clearly depicts a similar temperature trend as measured by the thermocouple at different reference temperatures. In figure 7(a), at room temperature, it is almost homogeneous throughout the spray whereas when the temperature of the injected liquid increases from $T_2$, $T_1$ and $T_3$, a clear difference in temperature gradients can be seen. As the liquid moves further down or sideways, its temperature drops slowly. In figure 7(b), a temperature field ranging between 28 °C to 30 °C is found. Similarly, in figure 7(c) temperature on the top of the spray is around 40 to 45 degrees while it decreases as droplets moves further down. In figure 7(d), droplet temperature is around 50 degrees near to the liquid injection point while near the spray tails edges and further downwards it reduces. It is important to mention that a large dynamic range of temperature difference is not apparent in the images because the droplets usually takes longer time to cool down in the range between 50°C to 20°C than in 100 °C to 50 °C.

5. Summary and Conclusions
A novel approach combining SLIPI and two-color LIF ratio is tested on a hollow cone water spray, using a Fluoresin dye as temperature sensitive material. It is found that the SLIPI detection scheme extracts a reliable intensity ratio with better dynamics of temperature gradient as a function of temperature than the conventional imaging approach. Due to discrepancies mainly arising from the multiple light scattering in CONV-LIF ratio, only SLIPI-LIF ratio is considered for calibration. Using this, a reliable two-dimensional map of droplet temperature is deduced. However, the accuracy of the method needs to be further investigated for a precise interpretation of temperature mapping in sprays.

Acknowledgements
The authors would like to thank Dr. Lars Zigan and Prof. Stefan Will from LTT Erlangen for co-supervising Stephanie Polster’s master thesis work. The Swedish Research Council is acknowledged for providing the financial support for the Project 2011-4272. Funding support from the European Research Council Advanced Grant DALDECS is also highly appreciated.

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