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# Instantaneous Temperature Imaging of Diffusion Flames Using Two-Line Atomic Fluorescence

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## Abstract

This work investigates the first demonstration of nonlinear regime two-line atomic fluorescence (NTLAF) thermometry in laminar non-premixed flames. The results show the expediency of the technique in the study of the reaction zone and reveals interesting findings about the indium atomisation process. Indium fluorescence is observed to be strongest at the flame-front, where the temperature exceeds 1000K. The uncertainty in the deduced temperature measurement is ~6%. The temperature profile across the reaction zone shows good agreement with laminar flame calculations. The advantages and inherent limitations of the technique are discussed.

# 1 Introduction

Temperature is a dominant parameter in a combustion system, both characterising the enthalpy of reaction and controlling many of the important chemical and physical processes. Laser diagnostics are the preferred tool for measurement of such parameters because they have the potential to provide *in-situ* non-intrusive, temporally and spatially precise information that is unrivalled by alternative methods. A variety of laser-based thermometry techniques have been developed [1]. However, most of these are limited to clean combustion environments and are not applicable in the presence of particles, such as dust, coal or biomass, and one of the most challenging, being soot. This limits the capacity to investigate and understand many systems of practical significance. The present paper addresses the need for laser-based thermometry in nonpremixed flames, with the intention to support future application in flames containing soot.

One of the most experimentally simple, yet useful, laser thermometry techniques is Rayleigh scattering [1]. Due to the elastic nature of the process, Rayleigh scattering is limited to very clean, particle-free situations. Filtered Rayleigh scattering has been developed to mitigate this interference [2–4]. The applicability of this technique in highly particle-laden flows has yet to be fully demonstrated. Raman-based thermometry has greater capacity to avoid problems with scattering interference, with Coherent Anti-Stokes Raman Spectroscopy (CARS) being one of the most widely used [5]. CARS is

more applicable to luminous and particle-laden flows than most other techniques. However, it has other limitations such as the necessity for line-of-sight access, experimental complexity, and a lack of spatial fidelity compared to planar techniques [1].

Amongst the laser spectroscopic techniques employed in combustion science, laser-induced fluorescence (LIF) is probably the most widely applied [1]. LIF can be used for temperature measurement, offering stronger signals than those from Raman systems. The temperature is deduced from the species' population according to the Boltzmann distribution. LIF thermometry has most commonly been employed to probe the *in-situ* flame radical OH, or additional NO seeded into the inlet stream [6–9]. However, the narrow range of temperature and mixture fraction over which OH exists in a flame limits its general application for thermometry [10]. Likewise, NO-LIF has limitations since it can suffer from background interferences in the presence of soot [9].

Two-Line Atomic Fluorescence (TLAF) is a laser thermometry technique based on the relative temperature-dependent population of two energy levels within an atomic species seeded into the flame. TLAF involves the sequential excitation and detection of the Stokes and Anti-Stokes transitions of a three-level system [11]. The temperature is subsequently deduced from the ratio of the two fluorescence signals. Not only does TLAF offer two-dimensional imaging, it has the added benefits of good sensitivity within a temperature range relevant to combustion and insensitivity to collisional quenching effects. In addition, the inelastic nature of the TLAF technique enables filtering

to minimize interferences from spurious scattering, allowing measurements to be performed in particle-laden environments. The advantages of TLAF over other laser-based thermometry techniques have been reported previously [1, 10–12].

Of the atomic species available, indium seeded into the flame has been identified as a suitable thermometry species for TLAF [13]. Indium has good sensitivity over the temperature range 800–2800K and both transition wavelengths are in the visible spectrum (*viz.* 410 & 450nm) where interferences are less pronounced than in the UV range.

Previous TLAF studies have been limited to low laser fluence in which the relationship between the excitation fluence and the resulting fluorescence signal is linear. Under such conditions, the signal is plagued by low signal-to-noise ratio (SNR), thus preventing useful single-shot imaging. Of the few studies that have applied TLAF, averaged results have typically been reported. With some notable exceptions (*e.g.* [14]) virtually all past work on TLAF has been performed in laminar flames, and thus the limitation of measuring time-averaged temperature has been acceptable. In the context of turbulent flames, however, such averaged data are insufficient.

With a view to improving the SNR of TLAF, the authors have sought to extend the technique into the non-linear excitation regime [11]. Non-linear regime two-line atomic fluorescence (NTLAF) was shown to provide superior signal and reduce single-shot uncertainty in premixed flames from  $\sim 250\text{K}$  for conventional TLAF to  $\sim 100\text{K}$ . The calibration constants were shown to

be independent of the fuel type and flame composition, within experimental uncertainty, across the stoichiometric envelope for a range of CH<sub>4</sub>, H<sub>2</sub> & C<sub>2</sub>H<sub>4</sub> premixed flames [11]. This independence from composition suggests that the technique will be applicable within the reaction zone of nonpremixed flames, however this is yet to be verified. The motivation of the current paper is therefore to assess the viability of the technique to cover the wider range of chemical compositions encountered in nonpremixed flames. Additionally, it is known that the local composition of the flame dramatically affects the fluorescence intensity due to changes in the mechanism of conversion of the indium chloride solution into indium atoms, leading to low signal in lean conditions [10–12]. This is a result of the requirement of the activation of the indium within the flame front [9]. For these reasons, the aim of the present investigation is to directly assess the performance of NTLAF in nonpremixed flames.

## 2 Methodology

The TLAF excitation/detection process consists of two distinct operations. The three energy levels of indium relevant to TLAF are shown in Figure 1. The Stokes process requires 410nm laser excitation ( $5^2P_{1/2} \rightarrow 6^2S_{1/2}$  transition), and the subsequent fluorescence is detected at 450nm ( $6^2S_{1/2} \rightarrow 5^2P_{3/2}$  transition). The Anti-Stokes process uses 450nm excitation ( $5^2P_{3/2} \rightarrow 6^2S_{1/2}$  transition) and 410nm detection ( $6^2S_{1/2} \rightarrow 5^2P_{1/2}$  transition). By adding a

small time delay between two excitation pulses, in conjunction with optical filtering, the Stokes fluorescence detected at 450nm may be essentially immune from spurious scatter from the 410 nm excitation. The same applies for the 450nm excitation and the 410nm detection. By both temporal and spectral shifting the Stokes and Anti-Stokes processes, measurements in sooting conditions are feasible.

A full description of the TLAF theory, especially incorporating the non-linear regime TLAF (NTLAF), has been presented in a previous publication [11]. In brief, for the NTLAF method, it has been shown that the temperature may be determined as follows:

$$T = \frac{\Delta E_{10}/k}{\ln\left(F_{21} \times \left(1 + \frac{C_S}{I_{20}}\right)\right) - \ln\left(F_{20} \times \left(1 + \frac{C_A}{I_{21}}\right)\right) + C_T}. \quad (1)$$

Here,  $\Delta E_{10}$  is the energy difference between levels,  $F$  is the fluorescence intensity, and  $I$  is the incident laser energy. The other terms are constants.  $C_A$  &  $C_S$  are derived experimentally from the fluorescence versus irradiance plot for the two excitation schemes, whilst  $C_T$  is determined via calibration in a premixed flame.

It has previously been shown that the calibration constants are independent of the fuel type and flame composition, within experimental uncertainty, for a range of premixed flames [11]. Given this independence from composition, it is anticipated that the technique will be applicable across the reaction zone of nonpremixed flames.

### 3 Experimental

The nonpremixed flames in this experiment are produced with the same flat-flame burner used for calibration purposes, and also used in previous work [11]. The burner consists of a series of tubes ( $\varnothing 1.6\text{mm}$ ) forming a 50mm square cross-section. For calibration, the burner is operated in a premixed mode, fuelled with a natural gas & air premixture. A portion of the air is passed through a seeder, consisting of an ultrasonic nebuliser, which generates a mist of  $5\mu\text{m}$  droplets of indium chloride dissolved in distilled water. The concentration of the indium chloride salt in the water was chosen to be 1.5 g/mL, which has previously been demonstrated to provide the optimal fluorescence signal, whilst minimising the potential affect of the metal salt on the soot formation process [11]. At this seeding level, the mass fraction of the water in the inlet gases is 5%. In the nonpremixed mode, the majority of the air flow is turned off, leaving sufficient air to seed the indium into the flame and also (for the present experiment) to clean the flame of soot. This corresponds to an air:fuel ratio of 2.3:1 by volume, thus exhibiting the same traits as a nonpremixed flame. The soot-free environment, while not necessary for the TLAf measurement, was chosen for the assessment of the present technique in nonpremixed flames, to eliminate any potential interference due to soot and to enable better modelling of the flame. The large cross-sectional area of the present burner results advantageously in a very low strain rate (flame time constant,  $D/U = 0.21\text{s}$  [15]), leading to a spatially



wide reaction zone.

The experimental arrangement used in this experiment follows that described previously [11]. In brief, two Nd:YAG pumped dye lasers are tuned to 410.18 & 451.13 nm and fired with  $\sim 100$ ns separation. The two beams are combined into a co-planar sheet of  $\sim 4.5$ mm thickness. The thick sheet allows a larger interaction region to be imaged and hence an improved SNR for the present study. Future experiments with turbulent flames will require a thinner sheet. The spectral irradiance of each of the sheets is  $2.5 \times 10^5$  W/cm<sup>2</sup>/cm<sup>-1</sup>, which has previously been shown to be in the nonlinear excitation regime [11]. The beams are directed through a cuvette in the same field of view as the burner. The cuvette is filled with fluorescing dye for the correction of laser energy variation across the sheet height and between laser pulses. The frequency-shifted fluorescence from both the cuvette and flame is detected through interference filters (centred at 410 & 450nm, 10nm bandwidth, 45% transmission) using  $f_{\#}1.4$  lenses onto two intensified CCD cameras. By appropriate image processing software, the resultant images from the two cameras are spatially matched using a three-point matching algorithm and then morphed based on the cross-correlation of a target to ensure sub-pixel spatial matching of the images. The in-plane spatial resolution of the matched images is 270  $\mu$ m. Measurements were conducted 10mm above the burner face. A schematic of the experimental layout is shown in Medwell *et al.* [11].

## 4 Results and Discussion

Figures 2(a) & 2(b) show typical instantaneous Stokes and Anti-Stokes fluorescence images, respectively, with the burner operating in nonpremixed mode. The flame-front is clearly indicated by the strong indium fluorescence. It is readily apparent that both the Stokes and Anti-Stokes transitions provide signal from approximately the same parts of the flame. This is due to the presence of sufficient concentration of neutral indium atoms in these regions. It is also evident that the Stokes signal has a higher SNR than the Anti-Stokes. The SNR (where noise is defined as the interpixel noise) of the images is  $\sim 20:1$  for Stokes and  $\sim 9:1$  for Anti-Stokes. Note that the images appearing in this paper have not been enhanced by image smoothing to reduce interpixel noise, as this may give misleading results regarding the SNR of the technique and also degrade the spatial resolution.

Figures 2(a) & 2(b) also indicate that no indium fluorescence is detected on either side of the reaction zone, implying a paucity of neutral indium atoms there. Whilst indium chloride is present in high concentrations along the jet centreline, the temperature and chemical conditions in this region are evidently not amenable to the formation of free, neutral atomic indium, as is required for TLAF. The mist generated by the nebuliser creates fine droplets of indium chloride dissolved in water. Upon heating near the flame-front, desolvation occurs whereby the solvent is evaporated, leaving the indium in the form of various metal complexes [16]. The desolvated ions and molecules

subsequently undergo a series of gas-phase reactions before neutral indium atoms are released [14]. The process by which the indium is converted into free atoms is dependent on numerous factors, one of which is the temperature. Away from the reaction zone the temperature is too low to generate sufficient indium atoms. Temperatures below  $\sim 800\text{K}$  also result in an Anti-Stokes ground-state population of less than  $\sim 3\%$ . The absence of fluorescence signal in the central region of the flame is a limitation. Nonetheless, data is obtained from the regions of greatest interest, notably in and around the reaction zone, where the temperature exceeds  $1000\text{K}$ .

It should also be noted that, for the present experiment, the surrounding air was not seeded with indium. While it has been shown that temperature may be deduced from TLAF under slightly lean conditions ( $\Phi \approx 0.8$ ) [11,17], the concentration of indium atoms, thus the intensity of its fluorescence, is known to be low under lean conditions [11]. Previous investigations have suggested this decrease is due to an abundance of oxidising species under such conditions [10,12]. Hence, seeding the surrounding air does not improve the level of signal there and would lead to additional cooling effects due to the presence of extra water solvent.

Applying the NTLAF theory to the fluorescence images yields the image of deduced temperature presented in Figure 2(c), which has a SNR of  $\sim 16:1$ . Due to this interpixel noise, the corresponding uncertainty in the temperature is  $\sim 120\text{K}$ , and is of this order over the entire temperature range. Those parts of the image for which no data is obtained are indicated to have a

temperature of below 800K. However, as described above, it is not possible to discriminate this temperature regime from other conditions not favourable for producing neutral indium atoms from the indium chloride. Nonetheless, from Figure 2(c) it is apparent that the temperature is obtained over a useful range across the reaction zone. Especially considering that understanding the coupling of temperature and soot is planned for future TLAFF studies, because soot is expected to exist only in the regions of the flames that TLAFF yields temperature, the lower-temperature limit of the TLAFF technique is quite acceptable.

Figure 3 presents the average temperature profile across the reaction zone over the range for which data can be obtained. This figure was produced by shifting each row of the temperature image (Figure 2(c)) to align the maximum temperatures and then taking the median average of each of the (20) constituent rows. This realignment of the temperature data is needed to account for the slight fluctuation in the flame-front location across the height of the image as the temperature data is sensitive to the steep spatial gradients which occur in nonpremixed flames.

Overlaid on Figure 3 is the temperature profile from laminar flame calculations of the current experimental conditions using the OPPDIF routine of Chemkin, with the GRI-Mech 3.0 mechanism. Due to the different configuration (coflowing versus counterflowing) the temperature profiles are not expected to match perfectly, but in general, good agreement is observed. The laminar flame calculations predict a peak temperature  $\sim 50\text{K}$  higher

than the measurements. A difference of this magnitude is expected since the cooling effects of the liquid-phase water solvent (calculated to be  $\sim 30\text{K}$ ) and radiation are not taken into consideration in the calculations, which in combination more than account for the  $50\text{K}$  discrepancy.

It is also apparent from Figure 3 that the lowest temperature recorded is  $\sim 1000\text{K}$ . This threshold is significantly higher than the  $800\text{K}$  required for the Anti-Stokes transition to yield a sufficient accessible ground-state population. This suggests that the indium formation process from the indium chloride solution does not become significant until  $\sim 1000\text{K}$ . This temperature limit is consistent with previous TLAF studies [11, 14].

## 5 Conclusion

In summary, the first demonstration of instantaneous temperature imaging in a laminar nonpremixed flame using nonlinear regime two-line atomic fluorescence (NTLAF) has been reported. Indium fluorescence is observed to be the strongest at the flame-front due to the local abundance of neutral indium atoms. This observation reveals important information on the formation of free indium atoms within the flame environment. Identification of the range where the NTLAF technique is applicable in nonpremixed flames using a water solvent to seed the indium highlights potential for further studies on alternative seeding arrangements. For the existing arrangement, across the reaction zone, where the temperature exceeds  $1000\text{K}$ , the uncertainty in

the measurement is found to be  $\sim 120\text{K}$ . The new-found knowledge therefore represents a significant development towards achieving two-dimensional thermometry in turbulent flames containing soot.

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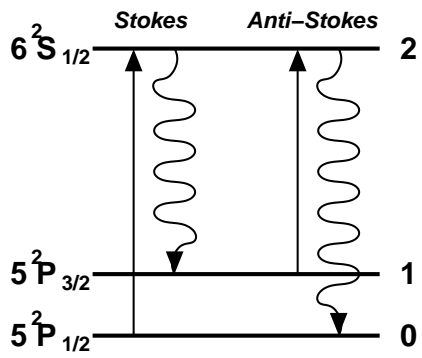
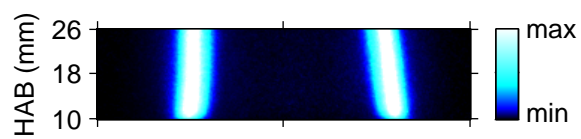
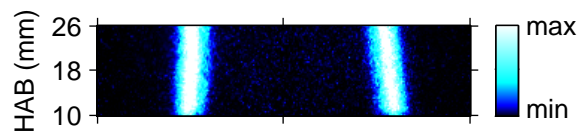


Figure 1: Indium atom energy transitions employed in Two-Line Atomic Fluorescence (TLAF).

(a) Stokes fluorescence



(b) Anti-Stokes fluorescence



(c) Temperature

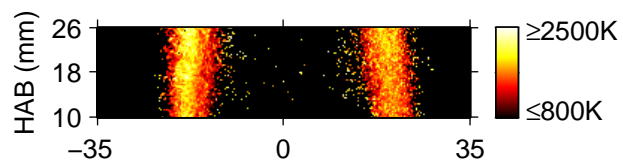


Figure 2: Typical instantaneous images of (a) Stokes and (b) Anti-Stokes fluorescence and (c) deduced temperature in a laminar nonpremixed flame. Image size approximately  $16 \times 70\text{mm}$ . HAB: height above burner.

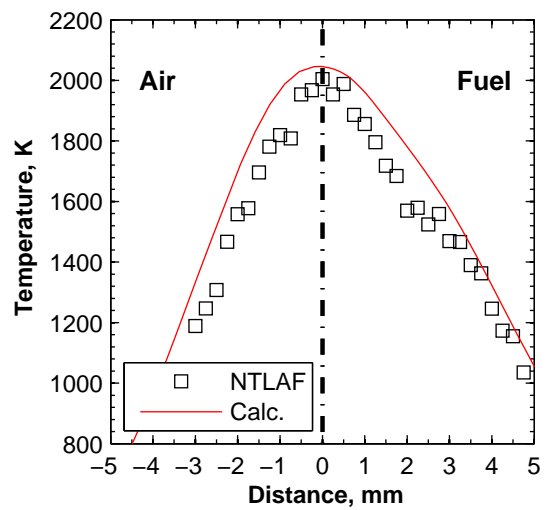


Figure 3: Temperature profile across reaction zone from NTLAF measurements and laminar flame calculations.

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