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Spectral Assessment of Interferences to Two-line Atomic Fluorescence (TLAF) in Turbulent Sooty Flames

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Abstract

Two-line Atomic Fluorescence (TLAF) using seeded atomic indium is a promising technique for two-dimensional temperature measurement in turbulent non-premixed sooty flames. However, it is desirable to reduce measurement uncertainties, which in turn depends on the fluorescence signal relative to background noise and interference. This paper reports a spectral assessment of interferences to both the Stokes and Anti-Stokes fluorescence signals in turbulent sooty flames undertaken with a spectrometer. Flame emission spectra were measured to assess the effects of various interference sources on indium fluorescence signals as well as to evaluate the efficacy of two techniques with potential to enhance the signal-to-noise ratio. Three types of interferences have been identified: laser Mie scattering from soot particles, laser-induced fluorescence from polycyclic aromatic hydrocarbons species (PAH-LIF) and laser-induced incandescence (LII) from soot particles. The feasibility of applying a correction for interferences by comparison of simultaneous images collected on- and off-resonance was assessed by collecting images at 430 nm and the fluorescence lines (410 and 450 nm). The correlation between the intensity of corresponding pixels from two instantaneous on- and off-resonance images was compared. This correction scheme was found to have limited value. The alternative scheme, which incorporates narrow-band filters, is considered to have much greater potential to be effective.

Keywords: combustion, laser diagnostics, 2D-temperature, turbulent sooty flame.

1. Introduction

Planar Two-line atomic fluorescence (TLAF) using seeded indium is a promising technique that can provide temporally and spatially resolved planar temperature information in turbulent sooty conditions [1,2] and in other particle-laden environments, in which the laser scattering by particles hinders the applications of other optical methods. In early application of the method, Kaminski *et al.* [3] reported measurements in an internal combustion engine. Engström *et al.* [4] and Nygren *et al.* [5] performed several studies on simple burner configurations. In these works, a low laser fluence was applied to achieve linear excitation regime indium LIF signals while keeping the laser induced emission from PAH or soot particles below a detectable level. More recently, Medwell *et al.* [6] extended TLAF into the non-linear excitation regime (NTLAF), i.e., using relatively high laser fluences to enhance the indium LIF signal and, thereby, to achieve a higher measurement precision [7,8].

At the high laser fluence typical of NTLAF, the interference of laser Mie scattering, LII from soot particles, and PAH-LIF may become significant relative to the indium fluorescence signals [8]. The diverse and complex nature of these interferences makes them difficult to account for by background subtraction. Chan *et al.* [9] assessed the interference signal by comparing radial profile of on- and off-wavelength measurements with LII measurements. They found that the interference, possibly from condensed species (CS) and PAH species, was significant and accounted for more than 50% of in some regions of these sooty flames, which is unacceptable. To broaden the range of applicability of the NTLAF technique, it is therefore important both to identify the sources of these interference and to seek efficient methods to reduce them. This requires detailed knowledge of the spectral characteristics of the interferences and of their temporal history relative to the excitation lasers, neither or which have previously been reported. The first aim of this paper is therefore to assess the time-resolved spectra of the aforementioned sources of interference to both the Stokes and Anti-Stokes fluorescence signals in turbulent sooty flames. The second aim is to assess the potential of various options with which to minimise each source of interference.

2. Experimental

Measurements were performed in a diffusion atmospheric-pressure ethylene-air jet flame issuing from a pipe (ID = 8 mm, OD = 9 mm). The flow rate of ethylene was kept at 6.5 standard litres (L_s)/ min and 13.5 L_s/ min for the air stream; resulting in a fuel equivalence ratio (Φ) of 6.9 and a jet exit Reynolds number of 3860. The flame was seeded with droplets of indium chloride dissolved in methanol at a concentration of 1.5 mg/mL. The droplets were produced in a sealed chamber with an ultrasonic nebuliser, and transported by the ethylene stream into the flame. Measurements were performed at a downstream location of 250 mm from the nozzle exit (x/d = 31.25), which is about half of the total flame length. Soot volume fraction has been measured previously in this flame [9]. At the measurement location, the soot volume fraction was determined to be

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~ 4 ppm by interpolating emission signal collected at 430 nm from the data.

The description of the optical arrangement for the laser system can be found in previous work [9] and is displayed in Fig. 1. The TLAF of indium employs two laser induced fluorescence processes based on the measurement of the Stokes and anti-Stokes direct line fluorescence of seeded indium. An alternate excitation and detection scheme is used to determine the temperature. The excitation wavelengths are 410.18 nm and 451.13 nm and the corresponding fluorescence wavelengths are collected around 450 and 410 nm, respectively. The excitation laser beams were performed alternatively with circular, horizontal and vertical polarisation.

A spectrometer (Andor Shamrock, SR-500i) combined with an ICCD camera (Andor iStar, DH734-18H-13) was used to record the laser-induced signals from flames. Spectra were accumulated from 100 instantaneous spectral images. Subtraction of dark charge and background as well as flame spontaneous emission was performed for each accumulated spectral image. Vertical binning was then applied. Corrections were applied for variation of quantum efficiency with wavelength for the ICCD camera.

Two ICCD camera heads (PCO, HFSC Pro) were used to simultaneously record single shot planar on- and off-resonance signals in flames. Off-resonance signals were collected using a 430 nm band-pass filter (Andover, FWHM = 10 nm), while fluorescence signals were collected using band-pass filters centred at 450 nm and 410 nm for Stokes process and anti-Stokes process, respectively (Andover, FWHM = 10 nm). Dark charge and background subtraction was performed for each image, followed by matching two corresponding on- and off-resonance images.

3. Results and discussion

3.1 Effects of laser Mie scattering

Figure 2 shows the spectrum of interference during collection of LIF of indium in the sooting flame. The spectra were recorded with the band-pass filters (450 nm for the Stokes process and 410 nm filter for the anti-Stokes process) in front of the spectrometer while no indium was seeded into the flame. The start of the camera gate was synchronised with the start of the laser pulse. It can be clearly seen that both the laser Mie scattering and the broadband laser-induced emission contribute interference to the TLAF signal.

The strength of the laser Mie scattering signal originating from soot at the excitation laser wavelength is several orders of magnitude stronger than the laser induced emission signal at the collection wavelength so that it leaks through the filter block band (OD ~ 3) and contributes to the total signal being collected. The magnitude of the laser Mie scattering signal is dependent on the polarisation of the laser beam [10]. By choosing a suitable polarisation for the excitation laser beams, the scattering signal can be reduced. Moreover, during the measurements, it was observed that the indium LIF signal excited by scattered laser photons is detectable using a laser power of 0.3 J/cm^2 for the Stokes process, working as a minor interference.

The effects of the polarisation of the laser beam on the laser Mie scattering signal from soot particles is shown in Fig. 2. For the Stokes process with a laser fluence ~0.3 J/cm^2, the ratio of scattered signal at 410 nm derived from the horizontally polarised beam to that derived from the circularly polarised beam was 0.21. The ratio between the scattered signal at 410 nm and the fluorescence at 450 nm is 0.038 and 0.17, for horizontally polarised beam and circularly polarised beam, respectively, as shown in Fig.1a. For the anti-Stokes process with a laser fluence ~0.25 J/cm^2, as shown in Fig. 1b, the ratio of the scattered signal at 450 nm derived from the circularly polarised beam to that from the vertically polarised beam is 0.38. The ratio between the scattered signal and fluorescence signal around 410 nm was found to be 0.77 and 1.96, for circularly polarised beam and vertically polarised beam, respectively. It is clear that the laser Mie scattering signal resulting from horizontally polarized beams is sufficiently weak to impose negligible interference on signals collected for TLAF in this way.

3.2 Effects of different interference sources

In addition to the laser Mie scattered signals, other interference sources also contribute to the TLAF measurement collected in Fig. 2, i.e., to the broadband measurement around 450 nm in Fig. 2a and around 410 nm in Fig. 2b. Laser-induced fluorescence from PAH and laser-induced incandescence from soot particles are possible sources responsible for these interferences at the detection spectral channels. It is therefore important to evaluate the effects of these interference sources on both the Stokes and anti-Stokes measurement.

Since PAH-LIF and LII signals have different spectral characteristics and temporal behaviour, time-resolved spectra were recorded to distinguish them. Fig. 3 presents the spectra recorded at 0ns, 30ns and 60ns after the laser pulse for both the Stokes and anti-Stokes processes. The gate width was kept constant at 30 ns for all measurements. The duration of the signal

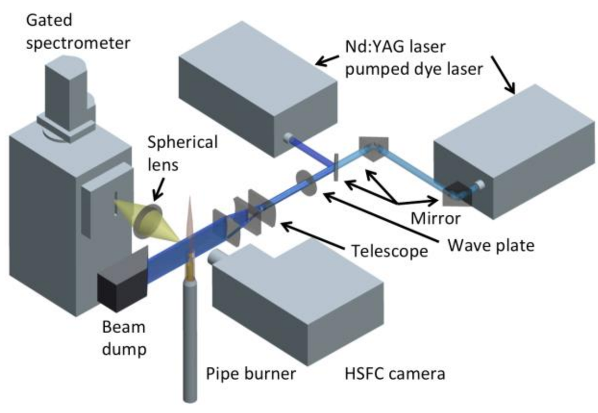
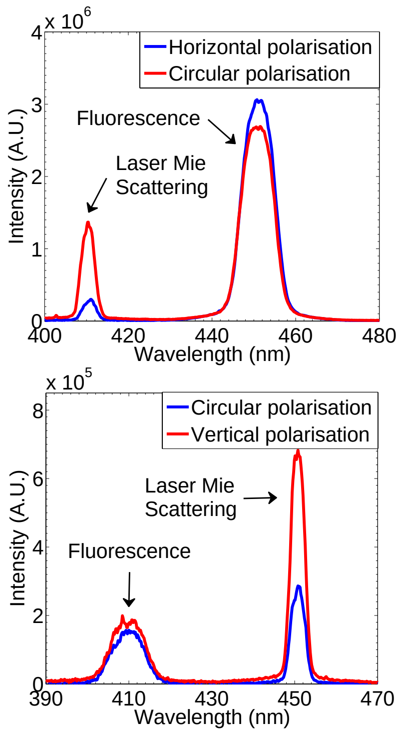


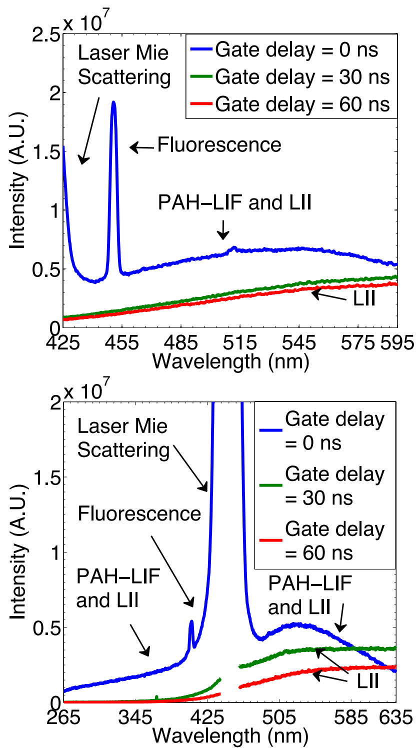
Figure 1: Schematic diagram of the experimental setup.



(a)

(b)

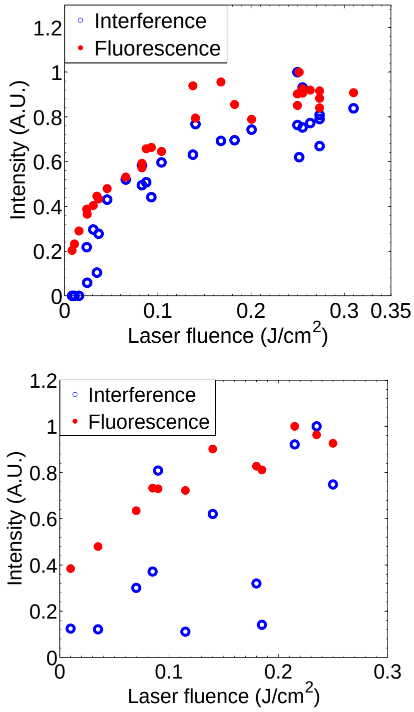
Figure 2: Spectral emission, recorded through a band-pass filter centered at (a) 450 nm for the Stokes process and (b) 410 nm for the anti-Stokes process. Spectral corrections were applied for variation of quantum efficiency with wavelength for the ICCD camera.



(a)

(b)

Figure 3: Broadband spectral emission with different delay times. No band-pass filter mounted for this set of measurements. Gate widths were kept constant at 30 ns for each measurement. (a) Stokes process; (b) anti-Stokes process.



(a)

(b)

Figure 4: Interference and fluorescence signals as a function of laser fluence. (a) Stokes process; (b) anti-Stokes process.

generated by laser Mie scattering matches that of the laser pulse and, while the fluorescence of indium and PAH is slightly longer, its lifetime is also of the same order. Therefore, these signals only contribute to the spectrum collected over the first 30ns, denoted t = 0ns. In contrast, the duration of the LII signal is of the order of 100ns due to the relatively long time required for the heated soot particles to cool down. Therefore, it can be deduced that the broadband signals observed in the spectra at 30ns and 60ns is attributable only to LII.

The broadband LII signal from heated soot particles is stronger at the longer wavelengths within the spectral range between 265 and 635 nm, as shown in Fig. 3. Therefore, the fluorescence signal collected at 410 nm for the anti-Stokes process suffers less interference fromthe LII signal than does that collected at 450 nm for the Stokes process. Moreover, at t = 0ns, both the Stokes and anti-Stokes fluorescence signals are affected more by interference from broadband PAH-LIF signals than by LII emission. However, the anti-Stokes signal is more susceptible to PAH-LIF than is the Stokes signal, as the ratio of fluorescence signal to PAH-LIF interference is much lower for the anti-Stokes transition than is that for the Stokes transition, as shown in Fig. 3. Importantly, since both are broad-band, the use of a narrow-band filter can be expected to efficiently reduce these interferences.

In addition, as shown in Fig. 3a, in the spectrum at t = 0ns, the weak peak around 510 nm is expected to be derived from laser-induced sublimation of C_2 species, which can be expected at these fluences.

3.3 Effects of laser fluence

The influence of laser fluence on the TLAF fluorescence and interference signals is presented in Fig. 4 as a function of laser fluence for both Stokes and anti-Stokes processes. These signals were obtained by integrating the spectral intensity centred at the 410 nm and 450 nm with a spectral width of 10 nm.

Figure 4 shows that the Stokes process reaches the non-linear excitation region at a laser fluence of ~ 0.05 J/cm^2, while the threshold for the anti-Stokes transition to non-linear excitation is less distinct but can be estimated to be around ~ 0.07 J/cm^2. The Stokes interference exhibits a similar behaviour to the fluorescence signal and is only slightly lower in magnitude within the nonlinear regime. The trends in anti-Stokes signal are broadly similar, although the interference exhibits much greater scatter. This scatter is attributed to instability in the mode of the laser. Significantly, Fig. 4 implies that the ratio of fluorescence to interference for both the Stokes and anti-Stokes processes are independent of laser fluence, for fluences above 0.05 and 0.07 J/cm^2 for the Stokes and anti-Stokes process, respectively.

3.4 Feasibility of on- and off-resonance detection correction scheme

By assessing the correlation between on- and off-resonance images, the feasibility of applying a detection correction scheme for interferences was evaluated.

It was found that the correlation between two on- and off-resonance images increases as the laser fluence increases. The average correlation coefficients obtained for 300 pairs of instantaneous images increases from 0.74 to 0.88 corresponding to an increase in laser fluence from 0.08 J/cm^2 to 0.32 J/cm^2. A stronger correlation was found for the Stokes process than the anti-Stokes process. At a fluence of 0.08 J/cm^2, the average correlation coefficient for 300 pairs of instantaneous images was found to be 0.74 and 0.59 for the Stokes and anti-Stokes process, respectively. However, even for the most correlated on- and off-resonance images acquired, the discrepancies were found to be unacceptable, which imply that, the uncertainty is still too high. In addition, both the on- and off-resonance images consist of complicated PAH-LIF, laser scattering and LII signals, the relative contribution of which is difficult to determine and varies throughout the flame. This makes the effects of subtraction of the two images difficult to interpret. For these reasons, the on- and off-resonance detection correction scheme is of little value.

4. Conclusion

Three types of interferences to the NTLAF fluorescence have been identified: laser Mie scattering from soot particles, laser-induced fluorescence by PAH species and laser-induced incandescence by soot particles. For both the 410 nm and 450 nm with a FWHM = 10 nm band-pass filter, Mie scattering can contribute up to 50 % of the total interferences (from PAH-LIF and LII and scattering). Importantly, changing the polarisation of the excitation laser beams from circular to horizontal can reduce this interference to negligible levels. The PAH-LIF interference is broadband for both the Stokes and anti-Stokes signals. It is difficult to reduce PAH-LIF interference by controlling the excitation laser fluence or to separate it from fluorescence signals by manipulating the detection timing scheme. However, the use of a narrow-band filter is expected to efficiently reduce this interference. It was also found that LII signal influences the Stokes signal more than the anti-Stokes signal because the LII emission is stronger at the longer wavelengths. The application of narrow-band-pass filters can also suppress this kind of interference.

The ratio of indium fluorescence to the interferences from PAH-LIF and LII near to the same wavelength was found to be independent of laser fluence for both Stokes and anti-Stokes processes.

The potential use of simultaneous on- and off-resonance collection to allow subtraction of the interference was found not to be effective for interference correction. This is because the interference is derived from multiple sources with different and complex dependences on both the excitation laser and on the local composition, in which the ratio of the sources of interference, such as PAH and soot, vary dramatically in a turbulent environment.

5. Acknowledgments

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6. References

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