Diode laser induced fluorescence spectroscopy for combustion thermometry

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Abstract. This paper describes the first use of novel blue diode lasers to make temperature measurements based on fluorescence. As a demonstration of this principle, indium atoms were seeded as a probe species into flames and the resulting diode laser induced fluorescence allowed an accurate determination of the temperature at a point. This permits spatially resolved measurements to be made, which are impossible to achieve using the established absorption based sensors. The technique opens up a range of application possibilities in dynamic and inhomogeneous reacting flows.

1. Introduction

The application of diode laser based sensors to industrial processes has been the subject of a substantial amount of research in recent years. Measurements have been made, for example of gas concentrations in the heavily particle-laden combustion chamber of a coal-fired power plant [1]. In another example, a methane sensor was implemented in a chemical vapour deposition reactor being used to deposit a thin film of TiO_2 on to a glass substrate [2]. Such applications were based on diode laser absorption spectroscopy. Similar diode laser absorption instruments can be used to perform temperature diagnostics in process flows but this line-of-sight technique only provides average temperature data in relatively homogeneous environments such as post-combustion exhaust gases [3], or unusual engine types with a one-dimensional temperature profile [4]. Many processes of industrial significance are characterised by substantial temperature inhomogeneities and fluctuations. A sensor that is capable of determining the temperature at a single point within the process would, therefore, be very useful.

This paper describes the development of a novel sensor that combines the practicality of diode lasers with the ability to make spatially resolved temperature measurements. This method works by using two blue diode lasers with wavelengths of 410 nm and 451 nm to probe the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ and $5^2P_{3/2} \rightarrow 6^2S_{1/2}$ transitions of indium atoms seeded to a flame at trace concentrations. The text is structured as follows: firstly a description of the operation of the sensor is given; this is followed by an outline of the experiments that were carried out to demonstrate its capabilities, a discussion of the results, and an overview of the future potential of this thermometry technique.

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2. Sensor Operation

Blue diode lasers [5] have become available fairly recently and represent a novel spectroscopic tool. In order to obtain tunable single mode radiation, a custom designed device (known as an extended cavity) had to be constructed around each of the Fabry-Pérot blue diode lasers, which are multi-mode in free-running operation. The principle of the extended cavity diode laser (ECDL) involves the use of narrow-band optical feedback from a grating [6]. It results in a laser beam with a very narrow line-width, which is therefore capable of high-resolution spectroscopy. This superior stability and spectral purity results in the potential for extremely accurate thermometry. The design of the blue ECDLs used in this project, and their favourable tuning properties, have been discussed in detail elsewhere [7].

The temperature sensor comprises two of these continuous wave blue ECDLs with wavelengths of 410 nm and 451 nm respectively. The two-line atomic fluorescence (TLAF) thermometry technique is based on laser induced fluorescence of indium atoms, which are seeded into the flame at trace levels (~100 ppb). Indium atoms are employed as a probe species because the oscillator strength of atoms is typically several orders of magnitude greater than that of molecules, so strong fluorescence signals can be obtained even with low power excitation sources. Atomic indium has a three-level energy structure (see Figure 1) consisting of a $5^2P_{1/2}$ ground state, an intermediate $5^2P_{3/2}$ state, and an upper $6^2S_{1/2}$ state. The lower two levels are closely spaced and their separation corresponds to an energy separation that is roughly equal to kT at flame conditions, which results in optimal temperature sensitivity. Previous TLAF studies have been carried out using argon ion laser pumped continuous-wave dye lasers [8], and Nd:YAG pumped dye lasers [9]. Diode lasers offer the potential for a compact instrument, which could replace the cumbersome systems used in the past.
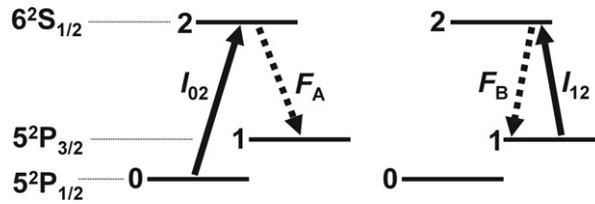
Figure 1. The excitation and detection scheme for TLAF.

The TLAF technique works by using the two lasers sequentially. A laser emitting near 410 nm is first used to excite the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ transition. The excited indium atoms will subsequently fluoresce by decaying back to both the $5^2P_{1/2}$ level and to the $5^2P_{3/2}$ level (which lies about 2220 cm^{-1} above the $5^2P_{1/2}$ ground state). Through the use of an appropriate filter in front of the detection system, only fluorescence at 451 nm (F_A) corresponding to $6^2S_{1/2} \rightarrow 5^2P_{3/2}$ is observed. Another laser emitting near 451 nm probes the $5^2P_{3/2} \rightarrow 6^2S_{1/2}$ transition and the resulting fluorescence at 451 nm (F_B) is observed by the same detector. The ratio of the fluorescence signals detected is related to the temperature by the following equation:

$$T = \frac{\Delta E/k}{ln\left(\frac{F_A/I_{02}}{F_B/I_{12}}\right) + 3ln\left(\frac{\lambda_{21}}{\lambda_{20}}\right) + ln\left(\frac{A_{21}}{A_{20}}\right)} \tag{1}$$

Here I_{12} and I_{02} are the laser irradiances at λ_{21} and λ_{20} respectively, ΔE is the energy separation between levels 0 and 1, k is Boltzmann's constant, and A_{20} and A_{21} are the Einstein coefficients for spontaneous emission. Further details of the theory behind the TLAF method can be found elsewhere [10].

3. Experimental method

Two blue diode lasers were purchased from Nichia Corporation, emitting at wavelengths around 410 nm and 451nm respectively. Diode laser injection current and temperature were controlled using commercial drivers (Tektronix). In order to obtain tunable single-mode output, a custom designed extended cavity was constructed around each diode. The output powers of the 410 nm and 451 nm ECDLs were 3.2 mW and 1.0 mW respectively. An etalon was used to determine the maximum single-mode wavelength tuning range that could be achieved with each laser without mode-hops.

Both were capable of tuning ranges of over 90 GHz, which exceeds the performance of commercially available systems.

The wide scans of laser wavelength are important in this application. In the case of fluorescence spectroscopy of an indium atomic beam [11], the spectral width of the individual transitions is very narrow. But the lines are substantially broadened by the conditions of atmospheric pressure and flame temperature [12]. Extended cavity diode lasers are typically characterised by very low spectral line-widths compared to many other types of lasers source. The line-width of the ECDLs used here was measured to be less than 6 MHz. This is more than a thousand times narrower than the pressure broadened indium spectrum and allows for very high-resolution spectroscopy to be performed.

Laser induced fluorescence spectra of indium atoms were recorded in a flame. The experiments were performed in a 10 mm diameter atmospheric pressure laminar CH_4/air Bunsen flame. The air stream was passed through a nebuliser containing an aqueous solution of $InCl_3$, resulting in a concentration of around 100 ppb of atomic indium in the flame. The output beam from the ECDL was focused to a diameter of around 100 μm in the flame. The fluorescence spectrum for the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ transition of indium at 410.1 nm was resolved by tuning the laser wavelength over a 75 GHz (2.5 cm^{-1}) wide region, at a scan repetition rate of 20 Hz. The resonant fluorescence signal originating from the laser focus was imaged at f/2.4 through a pin-hole of 150 μm diameter onto a photomultiplier tube (Hamamatsu, R3788). An interference filter centred around the indium fluorescence transition was used to suppress flame emission. Before entering the flame, part of the laser beam was split off by a glass plate and passed through a Fabry-Perot interferometer to monitor the relative wavelength change during the scan, and to ensure that the scan was mode-hop free. A separate photodiode was used continuously to monitor the laser power, which was used to normalise the acquired spectra. A similar procedure was followed with the other ECDL to probe the $5^2P_{3/2} \rightarrow 6^2S_{1/2}$ transition of indium at 451.1 nm.

Having acquired high-resolution fluorescence spectra, it was then possible to make temperature measurements in the same laminar flame. The two lasers were overlapped using a dichroic mirror and focussed to a point in the flame. The set-up for this experiment is shown in Figure 2. Sets of spectra were acquired sequentially with one laser then the other. In each case, the total fluorescence strength could be found by integrating the area under the acquired spectra, and the equation shown above was used to calculate the temperature. This was done at a range of positions along the central axis of the flame.
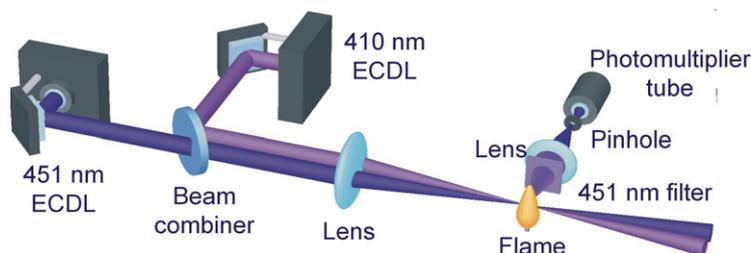
Figure 2. Optical set-up for two-line atomic fluorescence temperature measurements.

4. Results

The continuous single-mode tuning range of the ECDL systems allowed scanning of the laser wavelength over the entire width of both indium transitions. The acquired spectrum of the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ transition at 410 nm is shown in Figure 3, as an example. It consists of four hyperfine components, the positions and strengths of which are indicated in the figure. Shown are an average spectrum of 100 single scan spectra, a single scan spectrum obtained in 50 ms, and a theoretical fit of four Voigt profiles to the average spectrum. Relative hyperfine peak separations [11] and intensities [12] were specified in the fit.
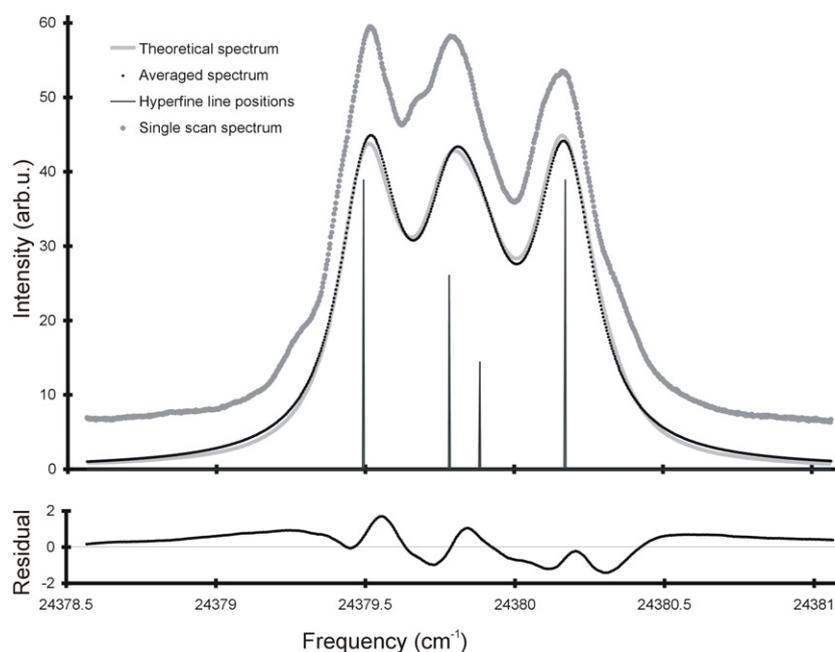
Figure 3. Indium laser induced fluorescence spectrum of the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ indium transition near 410 nm. A single scan spectrum is shown offset from the averaged spectrum to improve clarity. A theoretical fit to the averaged spectrum, the residual of the fit, and the positions and relative strengths of individual hyperfine peaks are also shown.

The fitted Voigt profiles yield a pressure broadening component of around 6.4 GHz, assuming a Doppler broadening component of around 2.2 GHz, corresponding to a temperature of 2000 K. The pressure broadening coefficient is in good agreement with that expected for N_2 dominated collisional broadening at this temperature and pressure [13]. Note that the total width of the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ transition is observed to be around 1 cm^{-1} (30 GHz), which is considerably wider than what has previously been assumed (2 GHz) and used in TLAF measurements [8]. This previous study was done using conventional dye laser systems whose spectral width is comparable to or even larger than the width of the indium transition that we now observe. This, therefore demonstrates the usefulness of these extremely narrow line-width (<6 MHz) blue ECDLs for high-resolution spectroscopy. Signal-to-noise ratios exceeding 150 were obtained even on single scan spectra, demonstrating the potential of the technique for measurements in dynamic systems.

Similar data were obtained for the $5^2P_{1/2} \rightarrow 6^2S_{1/2}$ transition using the 451 nm laser [14]. The total fluorescence signals were found by integrating the area under the spectra for each transition. Figure 4 shows vertical profiles of the 410 and 451 nm fluorescence signal levels along the centre axis of the flame. Data points correspond to the integrated fluorescence signal from 200 spectra, normalised by the laser power. At heights between 15.5 and 16 mm significant increases in the indium fluorescence signals are observed, as the neutral indium atoms are generated close to the flame front. The fluorescence signals then decrease gradually with increasing height, due to the oxidation of indium. Error bars shown correspond to the standard deviation of the integrated fluorescence powers obtained from single scans. Observed signal variations are thought to stem from dynamic changes in neutral indium formation. This is substantiated by the fact that error bars are largest near the tip of the flame where neutral indium production rates are strongly affected by minute flame flicker.
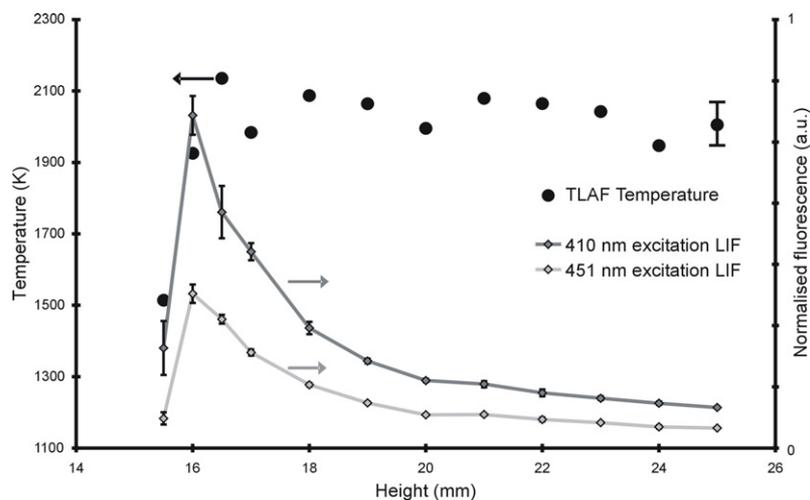
Fig. 4. Temperature and laser induced fluorescence (LIF) signals along a vertical axis through the centre of the flame.

The temperature profile, also shown in Fig. 4, was evaluated directly from averaged fluorescence powers using Eq. 2. The excellent spatial resolution demonstrates the superiority of TLAF over conventional line-of-sight absorption techniques to perform temperature measurement in spatially inhomogeneous combustion environments. A steep rise in temperature is seen near the flame front, with temperature staying nearly constant in the post-flame region, in agreement with previous studies of CH_4/air Bunsen flames [15]. Temperatures in the post combustion region are seen to fluctuate slightly from one measurement point to the next (around 2.5%). This scatter is a consequence of both minor drifts in local indium concentration and small flame movements due to currents of ambient air and flow speed variations. The error bar shown at h=25 mm reflects an upper estimate of the total systematic uncertainty.

5. Future prospects

The measurements reported in this paper have been performed in laminar flames. The viability of the TLAF blue diode laser thermometry technique has been confirmed by time-averaged measurements in this steady-state environment. The measurements were acquired with high signal-to-noise and the excellent spatial resolution distinguishes this method from anything that could be achieved with diode laser absorption spectroscopy. The usefulness of the temperature sensor would be further enhanced if it were capable of rapid measurements, thus permitting the study of dynamic process environments. An effort to dramatically increase the temporal resolution is presently being undertaken. We have a particular interest in turbulent combustion applications, and intend to make measurements in a running internal combustion engine. In order to resolve the fluctuations in such systems, it is typically necessary to execute individual temperature measurements in a time of less than one ten-thousandth of a second. A substantially different approach is required in order to speed up the measurements in this way; this essentially results from the fact that it is impossible for mechanical parts within the laser to move back and forth at such a rate. Some recent results have demonstrated the feasibility of performing ultra-rapid scans of the laser wavelength using a novel tuning scheme. This has the potential to allow high-speed temperature measurements in dynamic systems to be realised in the near future.

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