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Spatially resolved temperature measurements in a flame using laser-excited two-line atomic fluorescence and diode-array detection

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Spatially resolved temperature measurements in a CH₄-air flame have been achieved using two-line laser-induced fluorescence from indium atoms seeded to the flame.

Introduction

Recently there has been a large increase in the application of laser techniques for probing combustion processes. The most predominant techniques are Raman scattering, laser-induced fluorescence, and coherent anti-Stokes Raman spectroscopy (CARS), which have been described in several review articles and books (see, e.g., Refs. 1-3). It has been shown that laser-induced fluorescence can be used for space-resolved detection of OH radicals in flames⁴⁻⁶ and also for multispecies (OH, C₂) space-resolved flame detection.⁷

In this Letter it is shown how a space-resolved temperature measurement can be achieved by using atomic two-line fluorescence. The two-line-fluorescence technique has been used for point measurements of absolute temperatures employing lamps⁸⁻¹⁰ or lasers.^{11,12} In this technique the flame is seeded with atoms having a ground state and a metastable state separated by a few kilotesla. Atoms in both states can be laser excited to an upper level, from which the fluorescence light is detected to yield information on the population ratio of the lower states and thus the absolute temperature. Suitable atoms are, e.g., In, Ga, Tl, Pb, and Ba. In this experiment In has been used since the transition at 410 nm from the ground $5p\ ^2P_{1/2}$ state to the excited $6s\ ^2S_{1/2}$ state and the transition at 451 nm from the metastable $5p\ ^2P_{3/2}$ state to the excited state both lie in a spectral region well suited for dye lasers. In order to attain a high accuracy, a careful monitoring of laser intensity and line profile of the two transitions is necessary.

The method makes single-shot imaging temperature measurements feasible in principle. This necessitates the use of two lasers having well-controlled bandwidths. (Since our original intention was to make single-shot imaging temperature measurements, two pulsed dye lasers tuned to 410 and 451 nm, respectively, were used in parallel. One of the dye lasers, however, showed too large frequency instabilities to make precision measurements possible.) In our final version of the experiment, one stable dye laser was used, and the wavelength was shifted between the two transitions.

Normally in the two-line-fluorescence method, the detection is made on the wavelength not used for excitation, i.e., excitation at 410 nm and detection at 451 nm, and excitation at 451 nm and detection at 410 nm, respectively. Through this procedure stray light from the exciting laser can be suppressed, and further the oscillator strengths for the transitions do not enter the theoretical expression for the temperature. In our experiment stray light was negligible, and therefore we detected the fluorescence at 450 nm for both excitation wavelengths, which eliminated the changing of interference filters.

This scheme leads to a theoretical expression for the temperature, which is given by

$$T = \frac{\Delta E}{k} \left\{ 1 / \ln \left[\frac{F_1 L_2}{F_2 L_1} \frac{g_2(\nu_2) \eta_1(gf)_2}{g_1(\nu_1) \eta_2(gf)_1} \right] \right\},$$

where ΔE is the energy difference between the lower states and F and L are the photocurrents measured for the fluorescence and the laser intensity, respectively. $g(\nu)$ is the overall normalized line profile at laser frequency ν obtained when the laser is scanned over the atomic-absorption profile. η is the quantum efficiency of the detector, and gf is the multiplicity times oscillator strength of the transition. Indices 1 and 2 refer to excitation at 410 and 451 nm, respectively. Using experimental values¹³ of the gf factors

$$(gf)_1/(gf)_2 \approx 0.46$$

and our experimentally measured value of

$$g_2(\nu_2)/g_1(\nu_1) = 1.5$$

at line centers, we obtain the formula

$$T = 3183 \left[1 / \ln \left(\frac{F_1 L_2}{F_2 L_1} 1.96 \right) \right] \text{K},$$

which is the basis for our calculated temperature profiles.

Experiment

The experimental setup is shown in Fig. 1. The dye laser operating with stilbene 1 or stilbene 3 was pumped

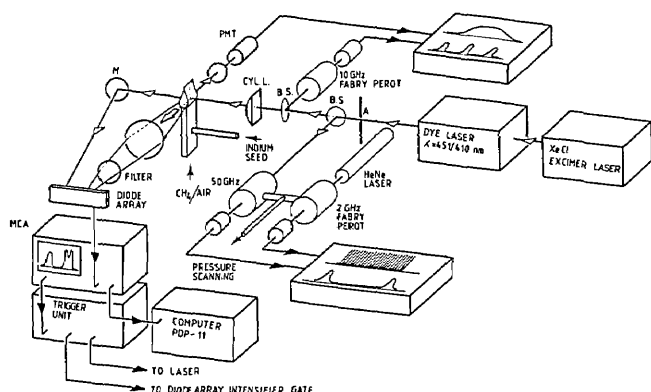


Fig. 1. Experimental setup used in the space-resolved two-line fluorescence temperature measurements. The gated electronics used for the spectral-distribution recordings are omitted for simplicity.

by a XeCl excimer laser (Lambda Physik Model FL 2002 and EMG Model 102, respectively) and was loosely focused into the flame with a lens.

The fluorescent light was imaged through a simple lens and an interference filter onto a diode-array detector (2.5 mm \times 25 mm; 1024 channels). A Tracor Northern Model TN-1710 IDARSS intensified diode-array rapid-scan spectrometer system, with an intensified and gated Model TN-1223-4IG detector, was used. The detector signals were fed to a multichannel analyzer, and the data were stored on floppy diskettes in a PDP-11 minicomputer for subsequent processing.

Simultaneously, the laser intensity was measured by splitting off a fraction of the laser light and directing it onto a few of the detector channels. Part of the light was transmitted through Fabry-Perot interferometers for determining the line profiles of the transitions and the laser.

Measurements

For the flame, a near-stoichiometric CH_4 -air mixture was used in an atomic-absorption premixed burner. A solution of 50 μg of In/ml was sucked into the predisperser of the burner through a capillary. By testing various concentrations from 10 $\mu\text{g}/\text{ml}$ to 1 mg/ml, we ensured that no effects of self-absorption were present at the concentration used. Further, the laser intensity was attenuated so that no noticeable saturation occurred. Because of the burner geometry, a cylindrical lens could be used to focus the laser beam, thus avoiding saturation while retaining maximum signal and spatial resolution. The line profiles for the transitions, $g_1(\nu_1)$ and $g_2(\nu_2)$, were determined by scanning the laser and recording the fluorescence signal together with interference fringes from a 10-GHz Fabry-Perot étalon. At 451 nm the half-width was 22 GHz, and at 410 nm it was 33 GHz, the main difference being due to different hyperfine-structure splittings since the laser bandwidth was only about 4–6 GHz for both wavelengths. From the resulting line profile the collisional broadening was estimated to be about 10 GHz.

In Fig. 2 the intensity profile obtained for one laser shot is shown, and in Fig. 3 the temperature profile as calculated from the data recorded by integrating over 100 + 100 shots 20 mm above the burner are presented. Taking into consideration the uncertainty in the determination of the fluorescence signal intensity, the laser intensity, and the line profiles, our estimated experimental accuracy is ± 200 K. Half of the uncertainty is attributed to the demonstration of the line profiles and slight drift of the laser across these. Any possible systematic error that is due to the uncertainty in the relative oscillator strengths has not been included. A 3% uncertainty (the accuracy stated in Ref. 13) corresponds to an additional error of 40 K. As can be seen from Fig. 3, the maximum temperature is about 2150 K, which is in good agreement with previous temperature measurements on a similar flame using CARS,¹⁴ yielding a temperature of 2125 ± 75 K. A calculated temperature for the burnt gases in a stoichiometric CH_4 -air flame is 2220 K.¹⁵

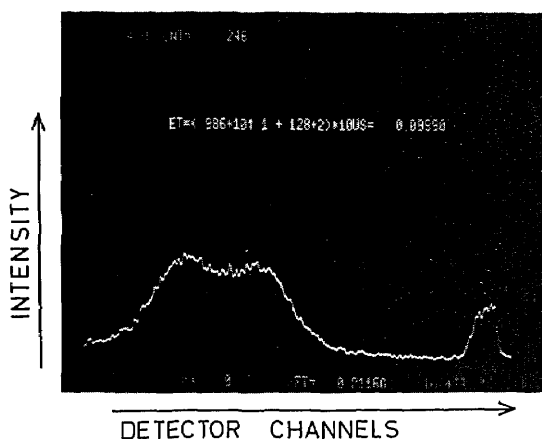


Fig. 2. A single-shot distribution of the fluorescence light at a height of 20 mm above the burner head when 410-nm laser excitation and detection at 450 nm are used. The peak to the right is used for laser-intensity normalization.

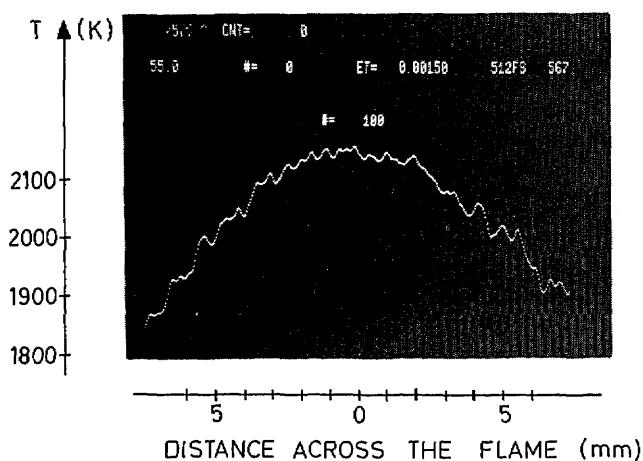


Fig. 3. Space-resolved temperature profile 30 mm above the burner head using 100 laser pulses from each 450–410-nm beam.

Discussion

Our results show that spatially resolved temperature measurements using laser-excited two-line fluorescence can easily be obtained, provided that no high time resolution is necessary. Further, the signal-to-noise ratio obtained for one single shot indicates that both space-resolved and time-resolved measurements are feasible when a second dye laser is used. In that case, the fluorescence light for each of the two wavelengths should be imaged through different interference filters⁷ onto two array detectors, which can be gated independently, and the lasers, both having well-controlled bandwidths, should be delayed at least 10 nsec with respect to each other, so that thermal equilibrium will be reached before the second pulse arrives. The time resolution would be further limited by the time to gate the detectors. Another approach, suggested in Ref. 16, is to separate the two laser beams slightly spatially and to image them onto a vidicon, which would increase the time resolution but decrease the spatial resolution.

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