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Single-pulse laser-induced OH fluorescence in an atmospheric flame, spatially resolved with a diode array detector

M. Aldén, H. Edner, G. Holmstedt, S. Svanberg, and T. Högberg

Laser-induced fluorescence measurements of OH have been performed in an atmospheric stoichiometric CH_4/air flame and in a highly sooting propane flame. The measurements were realized with a single 6-nsec pulse from a frequency-doubled dye laser pumped by a Nd:YAG laser and with a spatial resolution of ~25 μm. This was achieved by imaging through a suitably chosen filter set a section of the laser beam onto a gated and intensified diode array.

I. Introduction

During the last few years the great potential of laser spetroscopy techniques for probing combustion processes has been clearly demonstrated (see, e.g., Refs. 1–3). Laser-induced fluorescence, Raman and coherent Raman techniques can be used for measuring species concentration as well as temperature distributions in burning media. Whereas most initial experiments have been performed on clean nonluminous flames, applications to realistic, turbulent, and sooty flames are now being demonstrated.[4] A program of combustion diagnostics experiments was initiated in our group about three years ago and the results of our early experiments are discussed in Ref. 5. In this paper we report on a powerful application of laser-induced fluorescence techniques to the mapping of OH profiles in a CH_4/air flame even in the presence of large number of particles. An imaging technique using a diode array, viewing the laser beam crossing the flame through a properly chosen OH filter, was used for measurements with 25-μm spatial resolution using a single 6-nsec laser pulse.

Laser-induced fluorescence applied to the flame situation has been treated in papers by Daily *et al.*[6,7] The possibility to overcome many interpretation problems using saturated laser-induced fluorescence was pointed out by Baronavski and McDonald.[8] Point measurements of OH radicals in flames have been reported, e.g., in Refs. 9 and 10, and were also performed by our group.[11] OH is of fundamental interest in combustion processes as it is an important oxidant, entering a large number of the prominent flame chemical reactions.

Optical multichannel instrumentation is becoming increasingly important in applied spectroscopy. Normally, the array detector replaces the photographic plate in the focal plane of a spectrometer, enabling simultaneous recording of a full spectrum. This possibility allows single-shot registrations of detailed spectra, especially important in turbulent media applications (see, e.g., Ref. 12). However, the multichannel detector can also be used in a spatial imaging mode for simultaneous registration of a selected species in an appropriate spectral band. Concentration measurements of CH_4 in a jet using Raman spectroscopy[13] and temperature measurements using Rayleigh scattering[14] have been performed in this way. In this paper measurements of OH in a flame using such an imaging technique are demonstrated.

II. Experimental

The setup for the OH fluorescence measurements is shown in Fig. 1. We used a Quanta-Ray DCR-1A Nd:YAG laser, producing 700-mJ pulses at 1.064 μm with a repetition rate of 10 Hz. The pulse length was typically 6–7 nsec in the normal Q-switched operation. The infrared radiation was frequency doubled to 532 nm using a KD*P crystal. The green laser beam served as the pump source for a Quanta-Ray PDL-1 tunable dye laser. The pulse energy from the dye laser operating

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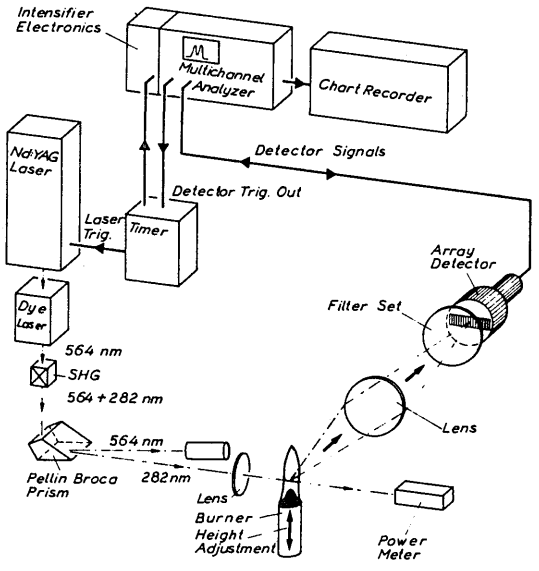

Fig. 1. Setup for the OH imaging experiments.

with rhodamine 6G was typically 100 mJ. To excite a ro–vibronic line in the $v'' = 0 \rightarrow v' = 1$ transition of the OH $A^2\Sigma \rightarrow X^2\Pi$ electronic band, the frequency of the dye laser beam was doubled using a second KD*P crystal. We excited the $Q_1(6)$ transition at 2829 Å, where the pulse energy typically was 20 mJ. With this high pulse energy we reached a substantial saturation which eliminates problems due to quenching, i.e., transitions due to collisions. Because of the saturation, which was verified by plotting the fluorescence signal vs the laser power, pulse-to-pulse energy fluctuations were not important. The UV beam was isolated from the dye fundamental beam with a Pellin-Broca prism and was focused in the flame with a quartz lens. A long focal length of the lens ($f = 100$ cm) was chosen so that a uniform cylindrical laser beam could be obtained through the 15-mm flame cross section. The burner used in the experiment with a stoichiometric CH_4/air flame was constructed so that the premixed gases were led through a 30-cm tube to assure a luminous gas flow. The terminal flame holder was water-cooled. The gas flows were measured with calibrated flowmeters. In the sooty flame experiments an ordinary Bunsen burner with propane was used.

The OH fluorescence light induced by the laser beam was isolated by means of a filter set consisting of two Schott colored-glass filters, WG 305 and UG 11. Clearly it is very important to suppress all spurious light such as Rayleigh and Mie scattering caused by the laser beam. The choice of proper filters was made in experiments where the detector array was placed in the focal plane of a Jobin-Yvon UFS-200 spectrometer with a dispersion of 24 nm/mm. This polychromator was used to record the spectral distribution of the light released from the intersection of the laser beam and the flame. The laser-induced fluorescence was imaged onto the detector array with an $f = 15$-cm quartz lens. We used a Tracor Northern TN-1710 DARSS (diode array rapid scan spectrometer) system with an intensified and gated model TN-1223-4IG detector. The minimum gating time of the 1024 diodes is 500 nsec, and the readout scan time is 10 μsec/channel. The spectral response of the detector array is that typical of diffused silicon photodiodes extending from ~200 to 1000 nm with peak sensitivity around 700 nm. The dimensions of the photodiodes are 25 μm × 2.5 mm. Thermoelectric cooling of the array detector gives a substantial reduction in dark current. The synchronization of the experiment was achieved by introducing proper delays between the firing of the laser flashlamps, the Q-switching, the gating of the detector intensifier, and the start of the readout scan. A special trigger unit was constructed for this purpose.

To correct for the nonuniform intensity response over the detector array a reference UV lamp was used. The lamp uniformly illuminated all the photodiodes giving a reference recording which served as a normalization for all the subsequent distributions. The normalization was automatically performed with a photometric processor module TN 1710-29. By mismatching the triggering of the laser and the gating of the detector array we also assured that no spontaneous flame emission contributed to the signal. This could also be checked by tuning the dye laser off the excitation line, resulting in no detected OH fluorescence.

III. Results and Discussion

In Fig. 2 spectra of the laser-induced light, taken with the detector array in the normal spectral mode, are shown. The full line shows the spectrum with no filter inserted, with two short-wavelength peaks at ~282 and 309 nm. The first peak is due to resonant OH fluorescence and elastic Rayleigh scattering, whereas the second one is caused by OH fluorescence. The peaks in the

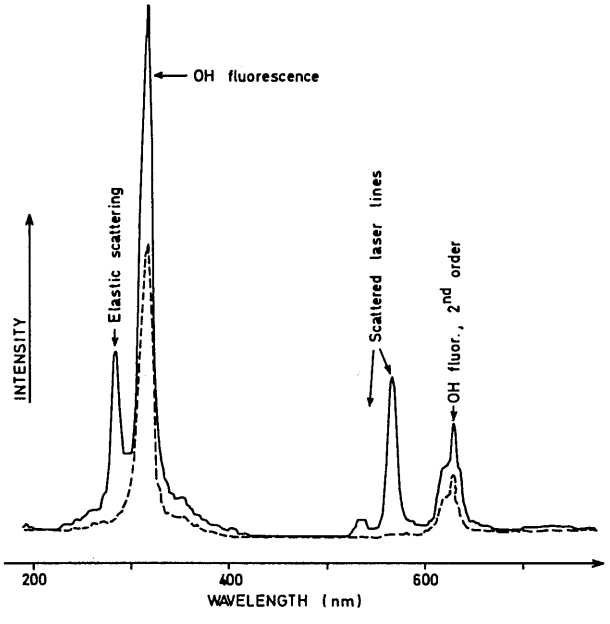

Fig. 2. Spectral distribution of the laser-induced light. The full curve is the spectrum recorded with no filters inserted, whereas the broken curve was obtained using the broadband filters.

green-yellow spectral region are due to scattered light from the doubled Nd:YAG laser (532 nm) and the dye laser (564 nm). At 620 nm OH fluorescence occurs in second order. With the filter set inserted the spectrum shown by a dashed line was obtained. From this spectrum it is apparent that the filter set suppressed all light but the OH fluorescence. Comparing the number of counts with and without the filters the transmission at the OH wavelength was estimated to ~60%. Both spectra in Fig. 2 were averaged over ten laser pulses. After using the detector array in the spectral mode the polychromator was removed and the spatial-imaging experiments were performed with the setup discussed in Sec. II. Results from the measurements on the stoichiometric CH_4/air flame are shown in Fig. 3, where every graph represents an OH distribution at a specific height above the burner. Each graph is obtained using a single laser pulse, and the data have been corrected for the nonuniform response of the photodiodes. Insert in Fig. 3 is a photograph of the conical flame. Since the flame was not shielded from the surrounding air the distributions at different heights are not exactly symmetrical because of flame flicker. In the same way the anomalously large peak reached at a height of 14 mm is due to flame instabilities, since the measurement where ten shots were averaged did not exhibit an enhanced intensity. It is interesting to notice the diffusion of OH molecules into the inner cone of the flame.

The applicability of the method to a luminous and sooty flame was demonstrated on a propane diffusion flame. The flame shown in Fig. 4(a) was bright yellow due to the presence of a large number of particles. These particles give rise to laser-induced fluorescence. In addition, polyaromatic hydrocarbons (PAH), present in the flame also yield fluorescence as has been reported by Coe and Steinfeld[15] and Fujiwara *et al.*[16] In Fig. 4(b), where recordings were taken with the laser tuned to the OH absorption line (upper curve) and with the laser tuned off that line (lower curve), the broadband PAH/particle contribution is clearly shown. In Fig. 4(c) the difference curve due to OH only is shown. Whereas OH is clearly measurable from such recordings, the background fluorescence strongly increased higher up in the flame resulting in small intensity difference between the on/off wavelength recordings. Since the on/off signals were not recorded simultaneously prob-

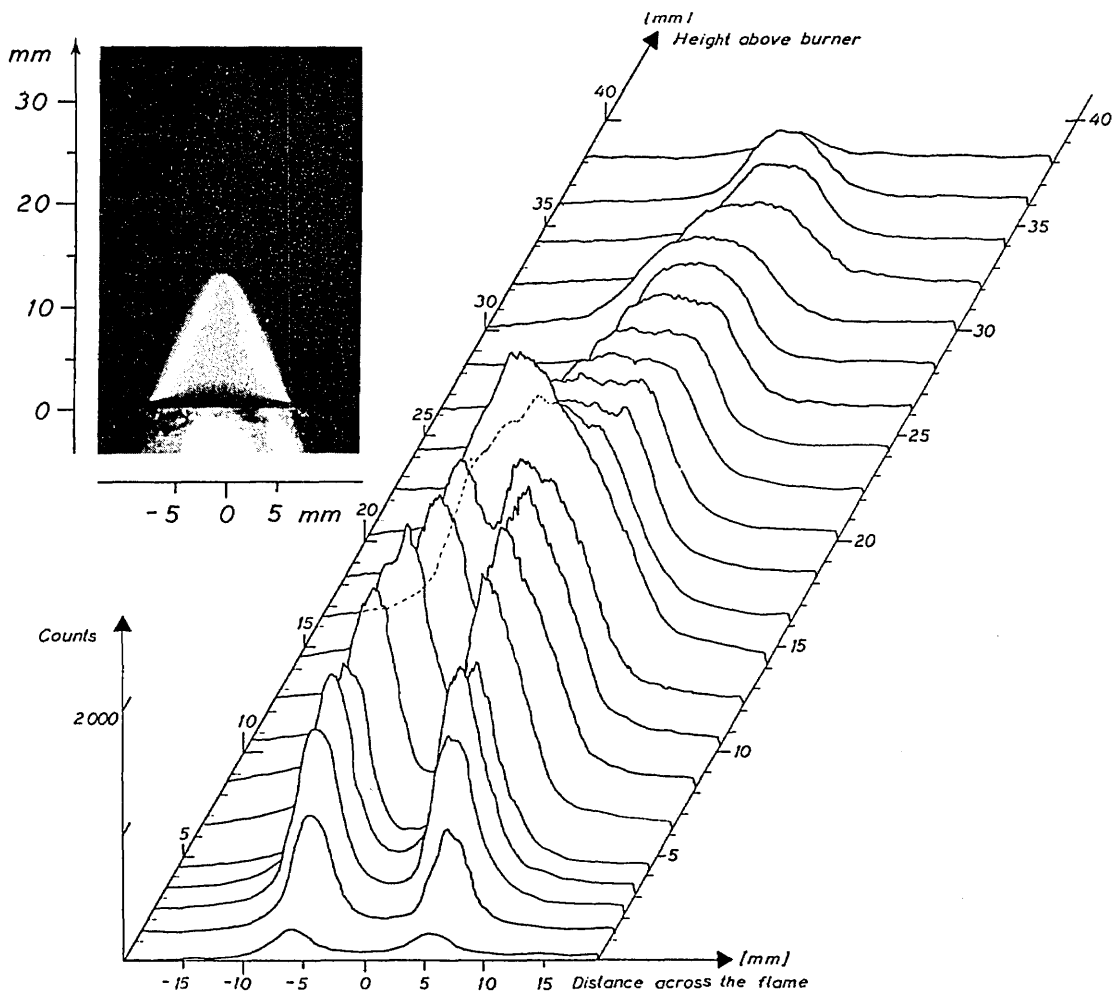

Fig. 3. Spatial distribution of the laser-induced OH light as a function of height above the burner. Insert is a photograph of the conical CH_4/air flame used in this experiment.

lems occur in the subtraction also in the lower part of the flame because of movements of the flame. The problems occurring with the broadband filter set could be eliminated by adding a narrowband interference filter (λ_{peak}:300 nm, $\Delta\lambda_{FWHM}$ = 11 nm). Although a strong reduction of the signal intensity occurred, it was then possible to measure the OH fluorescence even high up in the flame. In Figs. 4(d)–(f) such recording of OH fluorescence at three different heights are shown. The two curves in each photograph were obtained for OH on- and off-resonance conditions, respectively. Each distribution is an average over ten laser pulses. However, with an optimal filter, not available when the recordings were made, single-pulse measurements are clearly feasible.

So far no attempts have been made to determine the absolute concentrations of OH in the flame. Several effects have to be considered in detail in order to obtain absolute measurements. Clearly the temperature distribution will also substantially alter the relative distribution. The questions on the validity of the steady state condition during the laser pulse as treated by Lucht *et al.*[17] and on rotational redistribution during the pulse[18,19] have to be approached. However, theoretical calculations[20,21] suggest that an OH concentration of ~0.7% is reached in a stochiometric CH_4/air flame.

IV. Conclusions

The work presented in this paper shows that the laser-induced fluorescence technique combined with an optical multichannel detection system can be used for instantaneous (6-nsec) measurements of intermediate species such as OH in a flame with a spatial resolution of the order of a few microns. It is also demonstrated that measurements can be performed in highly sooty and luminous flames. We plan to use the technique in ignition experiments, studying the OH participation in that context. This type of measurement will be combined with CARS or spontaneous Raman techniques to measure the temperature evolution. The combination of these powerful techniques may enable an assessment of some of the important chemical reactions. Clearly the imaging technique demonstrated in this paper should also be applicable to other molecules of interest in combustion processes, e.g., C_2 and CH. Preliminary experiments on C_2 in a C_2H_2/O_2 flame have been performed and the results look very promising.

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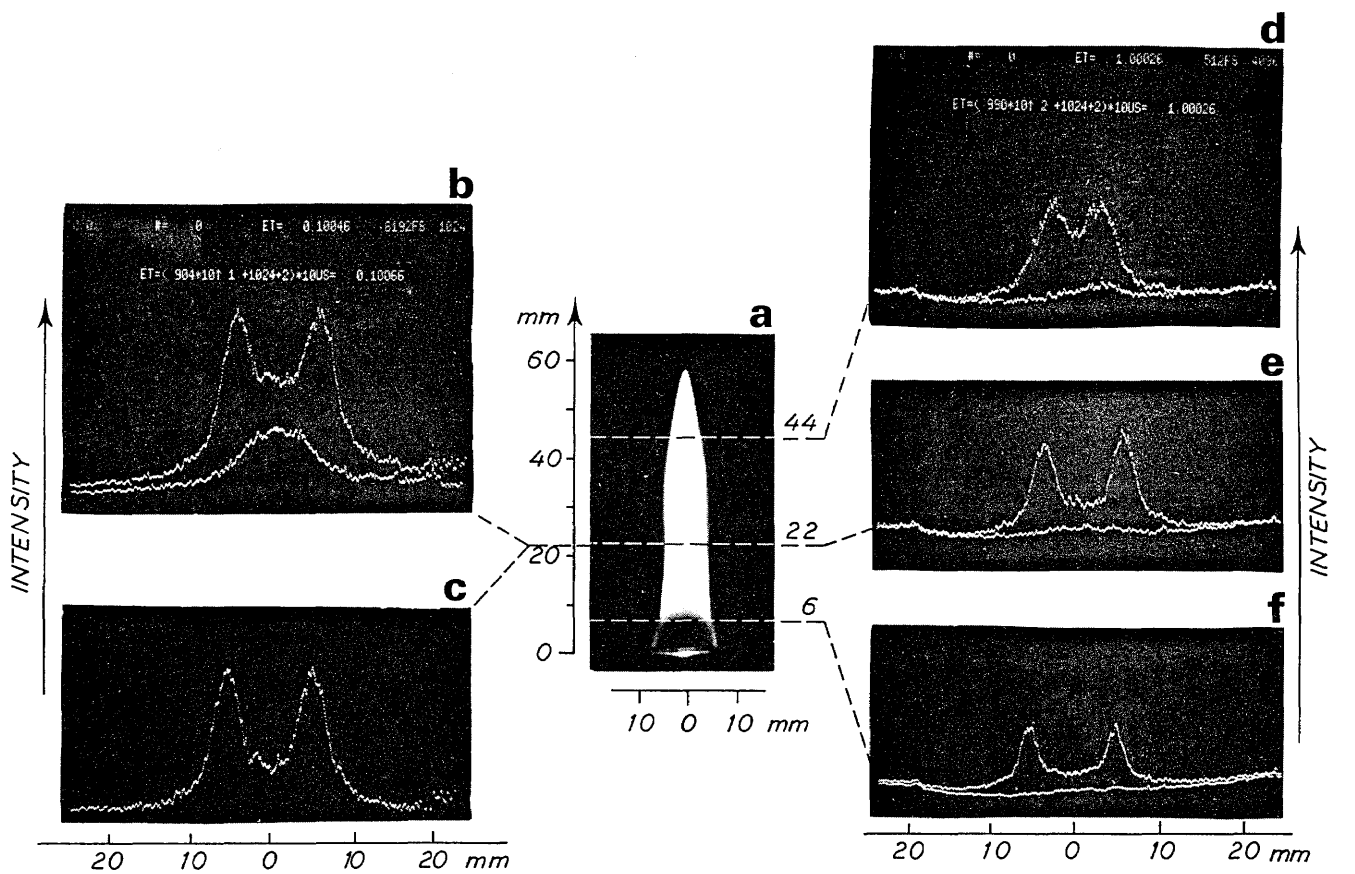

Fig. 4. (a) Flame used in the sooty flame experiment. (b) Distribution of the laser-induced light on/off OH resonance at a height of 22 mm (upper and lower curves, respectively). (c) Difference curve obtained from the data shown in (b). (d)–(f) OH emission distribution at three different heights above the burner, using an additional interference filter suppressing the particle/PAH laser-induced fluorescence. Each photo shows two distributions corresponding to on/off resonance.

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SPECIAL EVENTS

Much research activity in optics and astronomy is located in Tucson. Several of the centers of this activity will be available for organized and private visits. These include the Optical Sciences Center of the University of Arizona and the headquarters of the Kitt Peak Observatory, located at the University of Arizona. The rsearch and education program of the Optical Sciences Center spans the full spectrum of optical sciences. There is also optics related activity at local companies such as IBM and Hughes, and in nearby Phoenix at Motorola, and Arizona State University.

A variety of planned and informal tours to the many interesting attractions in the area are planned. These include the Arizona-Sonora Desert Museum which provides an admirable introduction to the plant and animal life of the Sonoran desert, the Kitt Peak Observatory, the Mount Hopkins Multiple Mirror Telescope, copper mines, Indian reservations, and, for an international flavor, Nogales, Mexico, 60 miles to the south. The city is ringed on three sides by mountains and there are many canyons available for hiking. The weather is normally dry with a temperature of 75-85 degrees. Nighttime lows are of the order of 55-65 degrees.

ABSTRACT DEADLINE

JUNE 11, 1982

TECHNICAL EXHIBIT:

An informal exhibit of table top displays of equipment and materials will be held in conjunction with the meeting. Ample time will be allowed for the attendees to visit the exhibits and exchange information with representatives of companies from all areas of the optical community. Information on the exhibit can be obtained from:

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