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Simultaneous multiple species detection in a flame using laser-induced fluorescence

Ulf Westblom and Marcus Aldén

An approach for simultaneous detection of NO, OH and O in flames using laser-induced fluorescence is presented. The technique is based on spectral coincidences using a Nd:YAG-based laser system producing a frequency-doubled and frequency-mixed laser beam at 287 and 226 nm, respectively. The possibility of making spatially resolved measurements using a diode-array detector was also investigated. The use of this technique for studying potential laser-induced disturbances was also demonstrated.

I. Introduction

The environmental situation and our growing awareness of limited energy resources have strongly encouraged research in the field of fundamental combustion processes during the last several years. Besides the use of more advanced computers for predictions and modeling of combustion phenomena, the introduction of different laser techniques for nonintrusive measurements with high temporal and spatial resolution has greatly improved the possibility for a deeper understanding of different combustion related phenomena, e.g., chemical kinetics, turbulent flows, ignition phenomena, and mechanisms for formation of air pollutants. Of numerous laser techniques that have emerged since the invention of the laser more than 30 years ago, three techniques have proven to be of special importance for diagnostics of combustion processes: Raman scattering (RS), coherent anti-Stokes Raman scattering (CARS), and laser-induced fluorescence (LIF).

The three techniques are complementary rather than competitive since Raman scattering is mostly used for measurements of major species and temperature in clean laboratory flames, whereas CARS is best used for temperature measurements in a highly particle-laden environment. The laser-induced fluorescence technique has the great advantage of being able

to measure low number densities of molecules, radicals and atoms preferable in laboratory flames. For different applications of these techniques for studies of combustion processes see, e.g., Refs. 1-3.

In the LIF technique a molecule, radical, or atom is excited from the ground state to an excited electronic state. After a short time period wavelength-specific fluorescence radiation is emitted when the species returns to its ground state. This technique is thus species specific in both the excitation and fluorescence wavelength, which reduces the possibility for spectral interferences, at least for small molecules. LIF has already been used for detection of flame radicals absorbing in the UV and visible region, e.g., CH, OH, CN and C₂.⁴⁻⁶ With the introduction of high-power solid-state lasers, the technique was also extended to include measurements of flame species absorbing in the VUV spectral region, e.g., O, H, and CO,⁷⁻⁹ by using multiphoton processes.

Of particular importance with the LIF technique is the possibility of making simultaneous multiple-point measurements, so-called imaging measurements. This technique was first demonstrated on OH radicals along a line using a diode-array detector,¹⁰ shortly followed by 2-D measurements utilizing a diode-matrix detector.^{11,12} The imaging technique has also been developed for temperature measurements,^{13,14} 3-D species detection,¹⁵ and, recently, flow-velocity measurements with high (10 ns) temporal resolution.¹⁶

Traditionally LIF is a single-species detection technique; however, there have been some multiple-species LIF experiments reported. NH and OH were measured using a Kr⁺ laser,¹⁷ and the same group also used an Ar⁺ laser for simultaneous measurements of CH, CN and NCO.¹⁸ Using two independent laser systems, simultaneous spatially resolved measurements of C₂

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and OH have also been reported.¹⁹ By combining one- and two-photon processes, detection of NO₂ and NO using a laser wavelength at 452 nm was demonstrated.²⁰ It has recently also been demonstrated how several flame species could be detected using laser-induced fluorescence with overlapping resonances.²¹ In this experiment, OH, NH, CH, and CN were simultaneously excited by the use of a laser wavelength at 312.22 nm. It was also reported how nonlaser-excited species could be indirectly excited through collisional energy transfer processes.²¹

In this paper we present experiments investigating the potential for simultaneous detection of NO, O, and OH. NO and O are excited around 226 nm, where these species have a one- and two-photon resonance, respectively. Since this wavelength was produced using a Nd:YAG-based laser system through frequency doubling of the dye-laser beam followed by frequency mixing with the residual 1.06 μm, a laser beam around 287 nm was also produced. Consequently, there is a possibility that this beam could be used for excitation of OH radicals, which have absorption bands in this spectral region. Investigations were made to determine the extent to which the spectral coincidences were large enough for simultaneous detection and how the laser linewidth could influence these effects. Experiments were also performed on imaging measurements, where the potential for recording single pulse images of NO, O, and OH in an H₂/N₂O flame were investigated. Finally, experiments were performed to determine whether potential laser-induced disturbances could be investigated using this technique.

II. Relevant Spectroscopy

Before starting the experiments, the spectroscopy relevant to simultaneous excitation of the three species, OH, NO, and O, was examined. The energy-level diagrams illustrating the relevant transitions in these species are shown in Fig. 1.

Oxygen atoms were first detected in a cell by Bischel *et al.*²² The two-photon atomic oxygen transition around 226 nm is between two ³P states and can therefore result in nine different fine-structure transitions. Two-photon dipole selection rules for the case ω₁ = ω₂ and π polarization in both beams, however, excludes the 0–1 and the 1–0 transitions resulting in seven allowed transitions.²³ These seven lines are usually not resolved since the energy splittings in the excited state, 0.54 cm⁻¹ and 0.16 cm⁻¹, respectively, are too small to be resolved under the combined effect of Doppler and collisional broadening and the broadening due to the finite bandwidth of the laser. In the ground state, the energy splittings are 158 cm⁻¹ and 226 cm⁻¹, respectively, resulting in a spectrum consisting of three resolved peaks of which the strongest (No. 1) consists of three unresolved fine-structure components and the following two (No. 2 and No. 3) each consists of two unresolved components. From the excited state the atom can decay either to the ³S₁ state giving fluorescence at 845 nm, or by collisional energy transfer populate the ⁵P_{1,2,3} state which then decays to the ⁵S₂

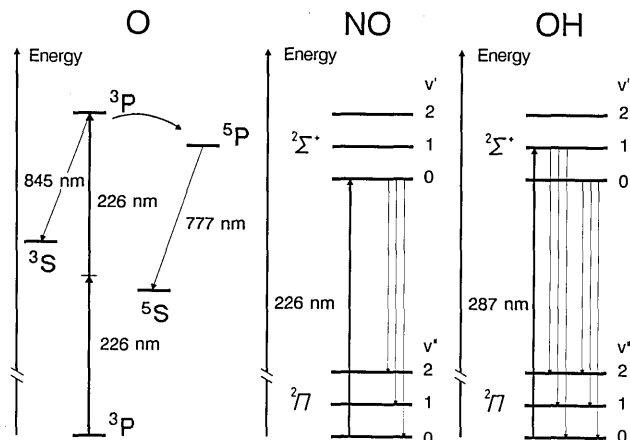


Fig. 1. Energy-level diagram illustrating the adequate transitions for simultaneous excitation of O, NO, and OH.

state giving fluorescence at 777 nm. The fluorescence emitted when the atom decays to its ground state is in the VUV and can thus not be used for flame detection of this species because of strong absorption in this wavelength region from other flame species.

For NO and OH, the situation is quite different. Not only are there different electronic states but also a large manifold of vibrational and rotational levels. The rotational transitions for these species that have the potential of overlapping with the two-photon oxygen lines are those that belong to the $A^2\Sigma^+ - X^2\Pi, v' = 0 - v'' = 0$, transition, the so-called γ band system at 227 nm for NO, and to the $A^2\Sigma^+ - X^2\Pi, v' = 1 - v'' = 0$ band system at 281 nm for OH. The wavelengths, 227 nm for NO and 281 nm for OH, refers to the position of their respective bandheads. Expressed in terms of the dye-laser fundamental beam they become 577 nm and 562 nm, respectively. Since the NO and OH bands are degraded in opposite directions, NO to the blue and OH to the red, there is a region around 574 nm corresponding to the atomic oxygen resonances in the UV region where they overlap. This is especially true in a flame where the high temperature moves the rotational population distribution towards higher rotational quantum numbers. Since the spectral density of rotational transitions is much higher in NO than in OH ($B_{NO} = 1.7 \text{ cm}^{-1}$, $B_{OH} = 18.9 \text{ cm}^{-1}$) it can be expected that finding a spectral coincidence between O and NO will pose a smaller problem than finding one for O and OH.

III. Experimental

The experimental setup used in the multiple-species LIF measurements is shown in Fig. 2 A Nd:YAG laser (Quantel YG 581-10) producing about 400 mJ in the frequency-doubled output at 532 nm was used to pump a dye laser (Quantel PDL-50) operating with a dye mixture of Rhodamine 590 and Rhodamine 610 to yield a maximum output of around 100 mJ at 574 nm in 10 ns pulses. The dye-laser beam was thereafter first frequency doubled to 287 nm, then frequency mixed to 226 nm with the residual IR output from the YAG laser

using KDP crystals for both processes. The resulting powers at 287 nm and at 226 nm were both about 5 mJ. (However, when the doubled beam was used without additional frequency mixing the output power in the doubled beam was about 15 mJ.) The laser power focused into the probe volume was measured by a Scientech (38-0101) power meter and could be set by a Newport Research continuously variable attenuator (model 935-10) to any value between 0 and 3 mJ without changing the beam properties and/or steering the beam. The frequency doubling and mixing crystals were mounted in a tracking system (Quantel UVT) enabling well-controlled wavelength scans of both the frequency-doubled and mixed-UV beams, retaining at least 80% of maximum laser power over the entire scan range, which was about 50 Å. The bandwidth of the YAG fundamental at 1.06 μm was 0.8 cm^{-1} during the experiment. The dye-laser bandwidth could be either 0.4 cm^{-1} or 0.08 cm^{-1} depending on whether a narrow bandwidth package, NBP, including a prism beam expander, was installed in the dye-laser oscillator or not. The experimental recordings were made using the dye laser in both modes in order to evaluate the effect of a change in laser line width. After the frequency-doubling and mixing crystals a Pellin Broca prism was used to spatially separate the different wavelengths. The UV beams were then sent counterpropagating through the measurement region, focused by $f = 1000$ mm lenses mounted on $x - y$ translators, enabling exact positioning of the foci relative to each other. The relatively long focal lengths used were motivated by the fact that it is possible to produce additional atomic oxygen from vibrationally excited molecular oxygen even at low laser power densities, (0.2 mJ, $f = 1000$ mm). This phenomena has been investigated by Goldsmith.²⁴

A Nikon quartz lens, $f = 105$ mm $f/4.5$, collected the laser-induced fluorescence and focused it either onto the entrance slit (200 μm) of a monochromator, mounted parallel with the laser beam, or, in the imaging mode, through suitable filters directly onto the photocathode of a diode-array detector (Tracor Northern model TN-6144, 700 channels). The monochromator (Jarrell Ash) was equipped with three interchangeable gratings of 150, 600, and 2400 lines/mm, which yielded dispersions of 24, 6, and 1.5 nm/mm, respectively. A photomultiplier tube (RCA 31034) connected to a boxcar averager (PARC model 4402) was used as detector for the monochromator. In the imaging mode, the diode-array detector was used for recording the spatially resolved fluorescence profiles of the different species across the flame. The monochromator, PM tube, and boxcar averager were used to record the excitation spectra. The laser-induced fluorescence spectra were also recorded spectrally resolved using the spectrometer equipped with the diode-array detector. Since the lense used is well corrected for chromatic aberrations there was no need for refocusing when changing the experimental setup from detecting fluorescence in the IR region from oxygen to recording fluorescence in the UV from OH and NO. The quality

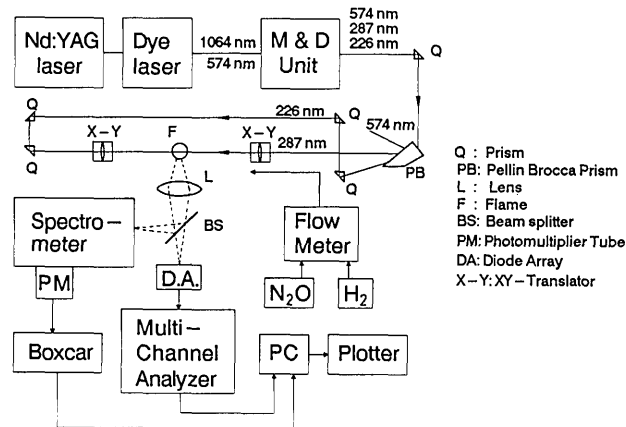


Fig. 2. Experimental setup used in the multiple species LIF experiments.

of the lens also made it possible to resolve spatial features of the intensity distribution across the flame with low image distortion. Thus, correct images could be achieved without applying the correction procedure described in Ref. 25. Correct focusing alignment was accomplished by replacing the flame with a thin (25 μm) electrically heated wire and then adjusting the focal distances to minimize the halfwidth of the wire image on the detector. The imaging ratio was 1:1.

Since the emphasis of the present work was to investigate spectral coincidences rather than to do quantitative measurements, a standard welding torch, operating at atmospheric pressure, was used as a burner. Hydrogen and nitrous oxide were used as fuel and oxidant, respectively, during most of the work. Acetylene/air and hydrogen/air flames were also investigated but all measurements reported here are from the $\text{H}_2/\text{N}_2\text{O}$ flame. The gas flows were controlled by electronic mass-flowmeters and the flame was kept fuel lean in all measurements, with an equivalence ratio of about 0.6.

IV. Spectroscopic Measurements

In Fig. 3 (a-c) excitation spectra of O, NO, and OH are shown where, since the linewidth at 1.06 μm was \sim a factor of ten larger than that for the dye laser (0.8 cm^{-1} compared to 0.08 cm^{-1}), the resolution in the OH recording is given by the line width of the dye laser and in the NO and O recordings by the Nd:YAG laser. The wavelength of the dye-laser beam was scanned over the same wavelength region for all three spectra, and they were recorded with one laser pulse per point. During these experiments, only one beam was present at a time in the probing volume to avoid potential cross effects (see below). The O spectrum, shown in Fig. 3(a), is two-photon induced, and shows the three resolved fine structure components of the $2^3P_{2,1,0} - 3^3P_{1,2,0}$ transition. In Fig. 3(b) the NO spectrum with the $A^2\Sigma^+ - X^2\Pi$, $v' = 0$ to $v'' = 0$, transitions in the γ band at around 226 nm is shown. The OH spectrum, with the $A^2\Sigma - X^2\Pi$, $v' = 0$ to $v'' = 1$ transitions around 287 nm, is shown in Fig. 3(c). Both these spectra are one-photon induced.

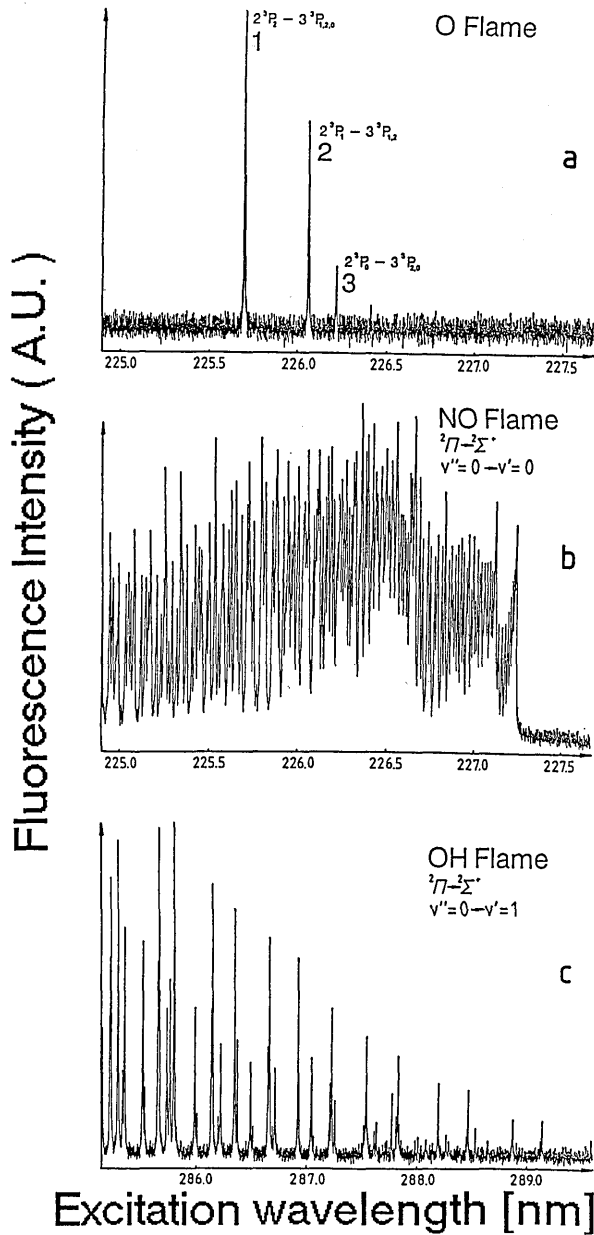


Fig. 3. Excitation spectra of (a) O, (b) NO, and (c) OH recorded from a H_2/N_2O flame.

To make a detailed investigation of possible spectral coincidences, the wavelength scale of the NO, OH, and O excitation spectra were overlapped and expanded around each of the three oxygen lines as illustrated in Fig. 4 (a-c). The solid lines represent the O profiles, the dashed, NO, and the dotted, OH. As can be seen in Figs. 4 (a) and 4 (b), which show the wavelength region around the two strongest oxygen lines, respectively, there is an overlap between NO and O and between NO and OH, but not between O and OH. Of course, an overlap can be achieved by making the laser linewidth sufficiently large but then the spectral power density has to be substantially sacrificed resulting in loss of signal, especially from oxygen atoms. (The effect of this can be seen in the recordings made in the imaging mode, which are shown both with the dye laser operat-

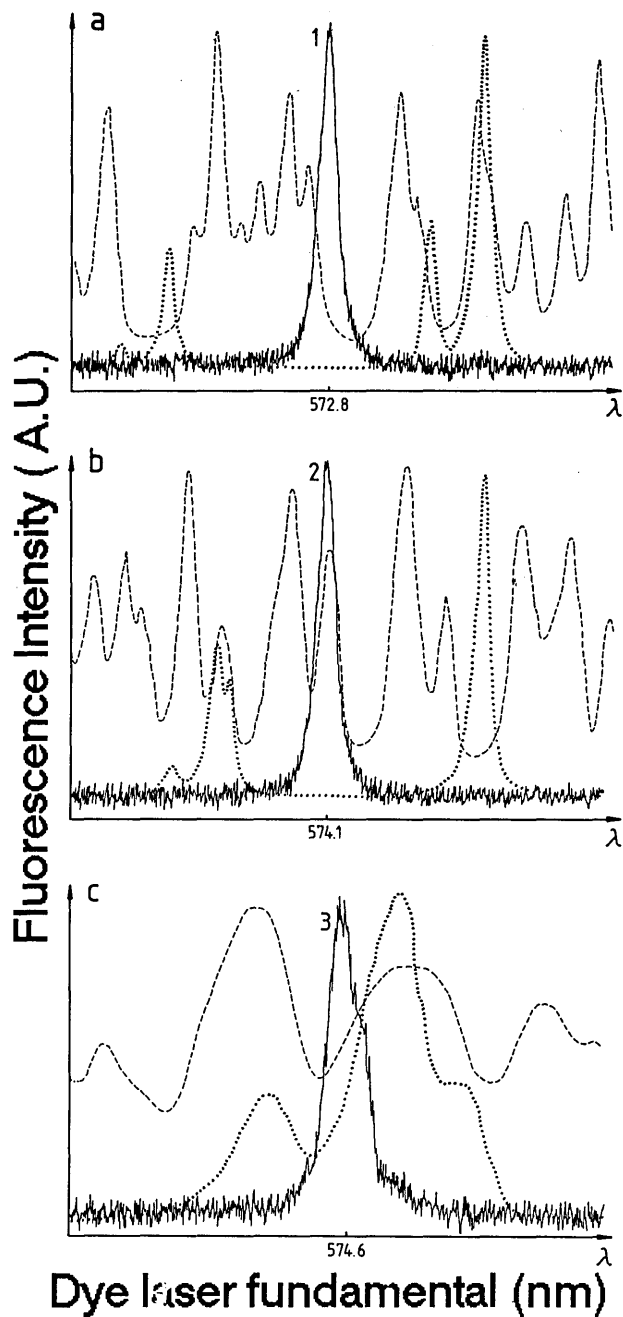


Fig. 4. Expanded wavelength scale of the excitation spectra of OH and NO around the three peaks of O (a) the $2^3P_2 - 3^3P_{1,2,0}$ transition at 225.7 nm, (b) the $2^3P_1 - 3^3P_{1,2}$ transition at 226.1 nm, and (c) the $3^3P_0 - 3^3P_{2,0}$ transition at 226.2 nm. [The wavelength scale is different in Figs. (a), (b) and (c)]: Solid lines = O; dotted lines = OH; dashed lines = NO.

ed broadband and narrowband as discussed below.) Consequently, since the distance to the nearest OH line is several times the laser linewidth both for the first and for the second O line, these transitions do not seem to be attractive alternatives for simultaneous excitation of both NO, OH and O. In Fig. 4 (c) the wavelength scale is further expanded around the third oxygen line and the spectrum is recorded with the dye laser operated in the broadband mode. As can be seen

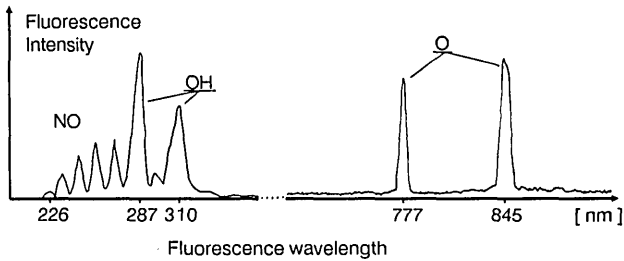


Fig. 5. Laser-induced fluorescence spectrum when exciting NO, OH, and O with a dye-laser fundamental wavelength at 574.85 nm.

there is here an overlap in wavelength for the three species which is easily achievable with standard Nd:YAG-pumped dye lasers.

Tuning the dye laser to the peak of the third oxygen line and resolving the emitted fluorescence through the spectrometer using the grating with the smallest dispersion, 240 Å/mm, we detected and recorded the radiation of the emission spectra with the diode-array detector as shown in Fig. 5. Since the spectrograph could not cover the entire wavelength range between 226 nm and 845 nm, the UV and IR fluorescence was recorded in sequence changing only the grating angle between the recordings.

V. Imaging Measurements

After the spectroscopic work described above was finished, the spectrograph was removed and the fluorescence was focused directly onto the diode-array detector through suitably chosen filters. In the case of NO an interference filter transmitting radiation around 260 nm ($\nu' = 0 - \nu'' = 3$) was used, whereas for OH an interference filter transmitting around 310 nm ($\nu' = 0 - \nu'' = 0$ and $\nu' = 1 - \nu'' = 1$) was chosen. For oxygen, a RG 9 3 mm Schott filter transmitting over 90% between 750 and 880 nm, was used. In Fig. 6 the spatially resolved fluorescence images of O, NO and OH are shown using the dye laser both in the narrow bandwidth mode (left column) and in the broad bandwidth mode (right column). For these recordings the dye laser was tuned to the strongest transition of oxygen. As was concluded above, the possibility for detection of OH radicals was limited here. The background seen in the OH recordings is caused mainly by flame luminescence and background radiation not suppressed by the gating (1 μ s gate) of the diode-array detector. The recordings acquired when the laser was tuned to the second oxygen peak are not shown since the results were similar to those shown in Fig. 6. However, if only NO and O are to be simultaneously detected, this is the oxygen transition having the largest overlap with NO.

In the recordings taken when the laser was tuned to the third oxygen peak, as shown in Fig. 7, where an overlap between all three species was found, it was seen that going from narrowband operation of the dye laser to broadband operation increased the OH fluorescence intensity by a factor of 10, while reducing the oxygen intensity by a factor of 3. The images show the spatial

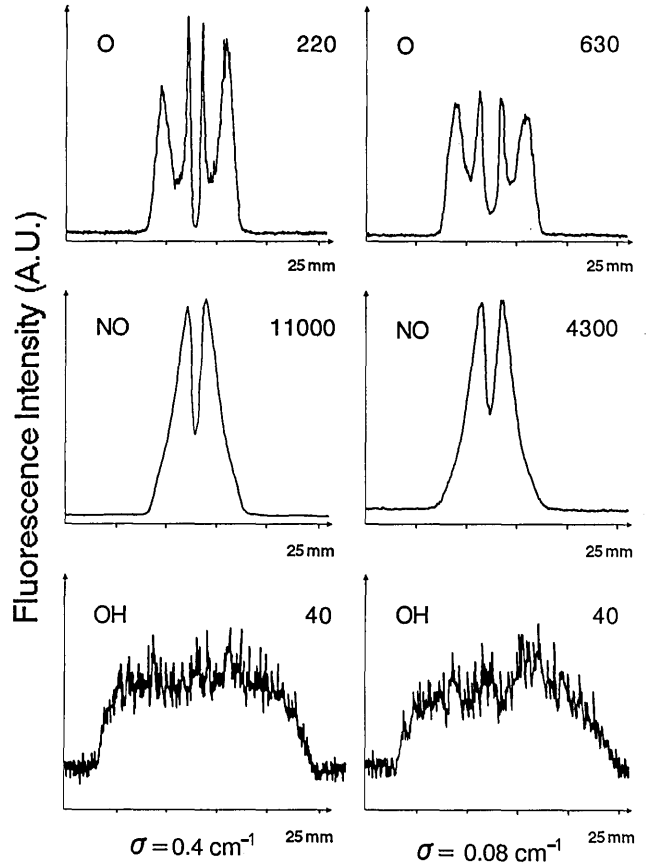


Fig. 6. Spatially resolved fluorescence distributions of NO, OH, and O with the laser tuned to the first oxygen peak. The distribution in the left column was recorded with the dye laser broadband (0.4 cm^{-1}) whereas the right column was recorded with a narrow band dye laser (0.08 cm^{-1}). The numbers shown in the figure indicate the number of counts at the peak.

intensity profile measured 5 mm above the burner head and through the reaction zone. For OH and NO there are two peaks indicating the position of the reaction zone and a dip in the middle where the fuel and oxidant are still unaffected. The nonzero concentration level in this area is mainly due to diffusion. For oxygen, the image is somewhat different; there is a first well-defined flame front, but also a second peak indicating a secondary reaction zone. The power levels used for the imaging recordings exceed those given in Ref. 24 for reliable quantitative measurements. However, the validity of the relative shape of the profiles was checked by comparing with recordings acquired with a laser power that was reduced by a factor of 10, and they were found to be almost identical. All recordings taken in the imaging mode were averaged over nine laser shots. They are not corrected for absorption which affects the symmetry of the profiles. As described in Ref. 26 this effect can be used for absolute concentration measurements when there is a reliable detected amount of absorption, which is normally a few percent when using pulsed lasers.

Investigations were also made to determine whether the technique described above could be used for measurements of laser-induced disturbances. We mea-

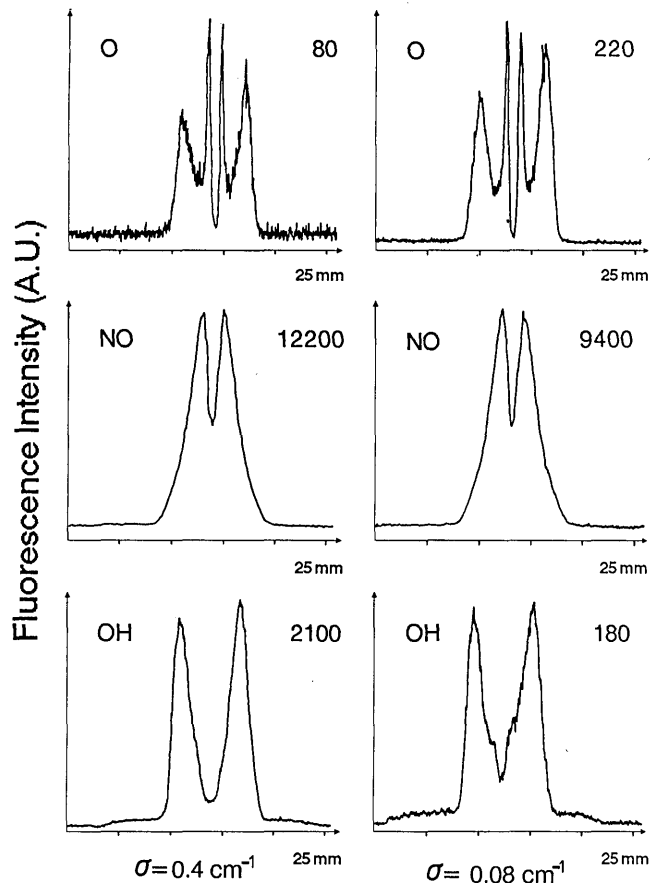


Fig. 7. Spatially resolved fluorescence distributions of NO, OH, and O when the laser was tuned to the third oxygen peak. The distribution in the left column was recorded with the dye laser broadband (0.4 cm^{-1}) whereas the right column was recorded with a narrow band dye laser (0.08 cm^{-1}). The numbers shown in the figure indicate the number of counts at the peak.

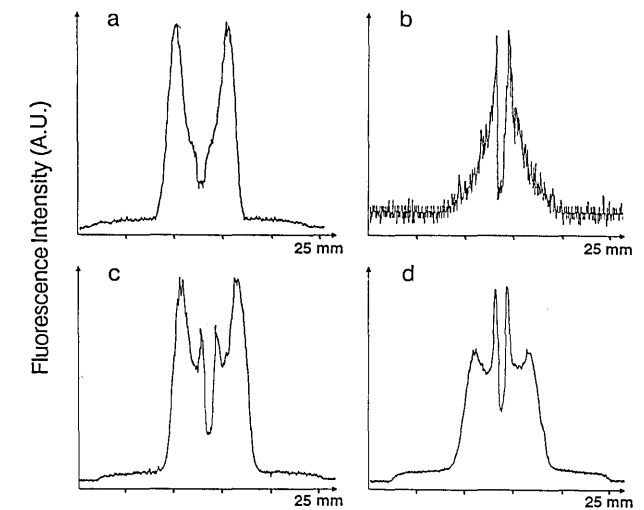


Fig. 8. Spatially resolved distributions as recorded through an interference filter at 310 nm. (a) With only the laser at 287 nm present, indicating the OH distribution. (b) With only the laser at 226 nm present, indicating the NO distribution. (c) The sum of the distributions in (a) and (b). (d) The distribution recorded experimentally with both laser beams present, indicating the presence of laser-induced disturbances.

present. If the profiles in Figs. 8 (a) and (b) are added, the result, shown in Fig. 8 (c), is expected to look like surements in principle require knowledge about this parameter. However, since the quenching is a function of pressure, temperature, and species composition in the probing point, a measurement of this quantity is a formidable task. One promising technique to get around the problem with quenching is to saturate the transition. This technique has proved to work very well for several flame species, e.g., OH.^{16,28}

During the course of this work we made preliminary experiments on the potential for saturation of the NO transition. The laser was tuned to the $R_1(22)$ transition in the NO ($0-0$) γ band and focused in the flame with a $f = 20\text{-cm}$ lens. It could then be seen that the fluorescence intensity curve as a function of laser intensity showed an appreciable amount of saturation behavior attributed to saturation, and thus in principle would permit absolute concentration measurements to be performed. Consequently, there will be a possibility for simultaneous concentration measurements of NO and OH by saturating the transitions. Further work in absolute concentration measurements is currently in progress in our laboratory. However, it should be noted that the power density required to saturate a transition in NO is apparently higher than what should be used for quantitative atomic oxygen measurements, without any laser-induced disturbances. In the case of making absolute number density measurements of oxygen atoms another approach has to be used: e.g., the calibration approach as proposed by Kohse-Höinghaus *et al.*²⁹

In this work three species could be excited simultaneously. However, as has been pointed out,³⁰ this number could actually be extended by another species, since at flame temperature, in the spectral region of

interest, it is also possible to excite O_2 in the Schumann-Runge bands. There are spectral coincidences between O_2 transitions and all three oxygen transitions,³¹ which thus might permit detection of four species, e.g., NO, OH, O, and O_2 , using only two single-laser pulses from one laser shot.

As indicated above there is always a probability of introducing errors in the measurements, especially when using high laser power in the UV spectral region. Thus, there have been several indications of such effects, e.g., C_2 formation in CARS³² and fluorescence¹⁹ measurements, appearance of artificial oxygen atom signals in room air³³ and also an artificially increased H and OH signal intensity when exciting hydrogen atoms at 205 nm.³⁴ Obviously great care has to be taken in order not to introduce errors in the measurements. These artifacts can not be unveiled only by going off resonance since the probed species can still be created although not detected. One technique which can be used to facilitate a correct measurement is a pump-probe technique, as described in Ref. 24, where a second laser system was used to probe the effect of too high laser power in measurements of O atoms in a flame. This technique was also used in Ref. 34.

Since a two-laser experiment requires a rather complex experimental setup, an alternative complement to this technique seems desirable. With the technique described in this paper it has been shown that under certain circumstances, it is possible to make a pump-probe experiment utilizing only a one-laser system.

We believe that the technique based on simultaneous excitation through spectral coincidences can be the profile acquired with both beams overlapping in space and time, which is shown in Fig. 8 (d). However, as can be seen when comparing Figs. 8 (c) and (d), there is an extra contribution in the recording acquired with both beams present. This effect was present both when the laser was tuned on and when tuned off resonance with oxygen. It was not possible however, to tune the wavelength off resonance with NO since the laser bandwidth was not narrow enough. The origin of the extra contribution to the inner peaks in Fig. 8 (d) has not yet been determined. The fact that it is only observed when both laser beams are present would seem to indicate that some form of photochemical perturbation in the flame is being induced by one of the laser beams (most likely the more energetic photons in the 226 nm laser beam), and then being probed by the other laser beam. This effect is currently being investigated in our laboratory.

VI. Discussion

As has been demonstrated in this paper it is possible to simultaneously detect NO, OH and O in a flame, using spectral coincidences. Also imaging recordings of these species are clearly possible.

In the work presented here the images were recorded sequentially by changing different filters by hand. However, by using a multispectral imaging device, e.g., as described in Ref. 27, simultaneously imaging recordings also seem quite feasible. The imaged record-

ings shown in this paper were all obtained by averaging over several laser pulses, while the spectra were recorded with one laser pulse per point. From the signal intensities during the spectral measurements, i.e., the spectra shown in Fig. 3, it was evident that the species could be detected simultaneously even using a single laser pulse. In the imaging experiment a trade off between large linewidth (to assure spectral coincidence) and high spectral brightness has to be made in order to make simultaneous single-shot recordings of all species. The spectral brightness is of special importance in the two-photon experiment in measurement of oxygen atoms. It is, however, clear that with the linewidth used in our experiments, out of NO, OH and O, any combination of two species, except O and OH, can easily be detected spatially resolved using a single laser pulse. The reason for this is simply that the NO resonances are so close that, with the laser linewidth used in this experiment and in the spectral region of interest, there is always a probability for excitation of NO as can be seen in Fig. 4 (a-c).

In this work no attempt was made to make quantitative measurements. There are two main problems with converting fluorescence distributions (like those in Figs. 6 and 7) to absolute number densities. First, the spectral coincidence for simultaneous excitation of all species is limited to certain rotational quantum numbers. Therefore, for NO the $Q_1(6)$ and $P_1(14)$ (which are unresolved) and $P_2(10)$ for OH, one cannot choose a rotational transition which is almost insensitive for temperature changes in the temperature interval of interest to avoid temperature effects in the species distributions. In our case this means that, e.g., for OH one would expect about a factor of 2 in population difference when going from 1000 K to 2000 K.

Second, since excited species suffer from quenching, i.e., nonradiating transitions, quantitative LIF measured to determine when not to use too high a laser power, although the technique can only indicate the presence of a perturbation and not the absence of such effects. It is also clear despite the fact that temperature and quenching effects limit the possibility for absolute number density determinations, that the technique could be of great value in showing trends and especially for detection of changes, for example, when introducing additives in a flame.

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