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Imaging laser-induced fluorescence of oxygen atoms in a flame

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During the last years different laser spectroscopical techniques have proved to be of great value in the diagnostics of combustion processes.[1] One of the most promising techniques is laser-induced fluorescence (LIF), since it offers high sensitivity and gives the possibility of spatially resolved measurements. The first spatially resolved measurements of a flame radical (OH) using LIF were performed on a 1-D diode array[2] followed by 2-D detection using a vidicon[3] and a diode matrix.[4] Contemporaneously with the development of imaging techniques, LIF has been extended to measurements of flame species that absorb in the VUV spectral region using multiphoton absorption processes. The first flame measurement of this kind with optical detection was performed on atomic oxygen[5] followed by H detection.[6,7] These species have also been studied by optogalvanic detection.[8]

Recently the imaging and multiphoton absorption techniques were combined and demonstrated on CO in a flame.[9] In this Letter we demonstrate spatially resolved detection of atomic oxygen in an atmospheric pressure C_2H_2/O_2 flame using absorption of two photons at 226 nm.

The experimental setup used in this experiment was very similar to the arrangement for two-photon CO monitoring described in Ref. 9. Since the setup is extensively treated in this reference only a brief summary will be given here.

A pulsed Quanta Ray DCR-2A Nd:YAG laser was used as a pump source for a Quanta Ray PDL-2 dye laser. With a mixture of approximately equal parts of rhodamine 610 and rhodamine 590 in the oscillator dye cell and rhodamine 590 tuned with hydrochloric acid in the amplifier cell, the output energy from the dye laser reached 80 mJ/pulse at 572.6 nm. Using KD*P crystals this wavelength was first doubled and then mixed with the residual 1.06-μm Nd:YAG fundamental beam yielding 1–2 mJ/pulse at 226 nm. A Pellin-Broca prism separated the beams of different wavelengths before the 226-nm beam was focused into an atmospheric acetylene–oxygen flame by a quartz lens. The fluorescence light was collected with an ordinary $f/2.8$ camera lens at right angle to the laser beam and focused onto the slit of a UFS 200 Jobin-Yvon spectrograph. A Tracor Northern diode array detector system (TN-1710 IDARSS) connected to the spectrograph monitored the spectra. In the imaging experiments the spectrograph was removed from the diode array, which then was used separately in a horizontal position. The synchronization of the laser and the detector gate was governed by a master trigger unit.

The excitation and detection scheme used in this experiment is schematically depicted in Fig. 1. Using the two-photon transition at 226 nm the $2p^33p\ ^3P_{0,1,2}$ state was populated followed by detection in the near IR. Oxygen atoms in this state either fluoresce directly to the $2p^33s\ ^3S_1$ state emitting 845-nm radiation or transfer through collisions to the $2p^33p\ ^5P_{1,2,3}$ state followed by 777-nm fluorescence, as observed by Miziolek.[10] The two-photon transitions to the individual fine-structure components of the ^3P state were induced separately. In our measurements we generally used

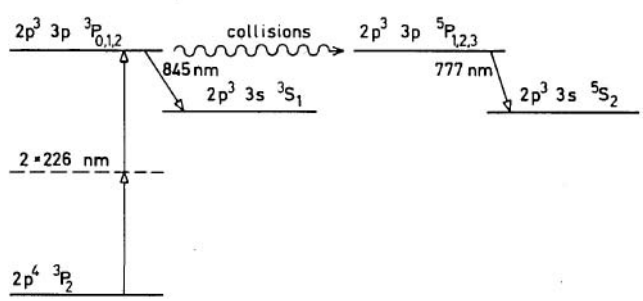

Fig. 1. Excitation and detection schemes for optical detection of O-atoms.

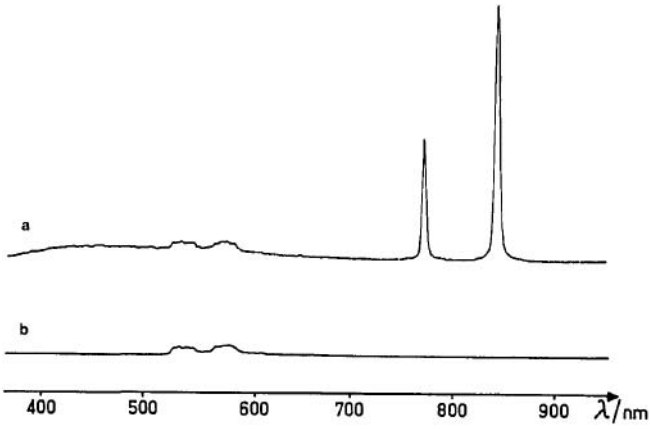

Fig. 2. *a*, Two-photon-induced fluorescence spectra from oxygen atoms in a fuel-lean acetylene–oxygen flame showing the triplet and the collision-induced quintet transitions at 845 and 777 nm, respectively. For comparison a spectrum from room air is shown in *b*. The broad peaks in the green-yellow spectral region are due to diffusely scattered laser light at 532 and 573 nm.

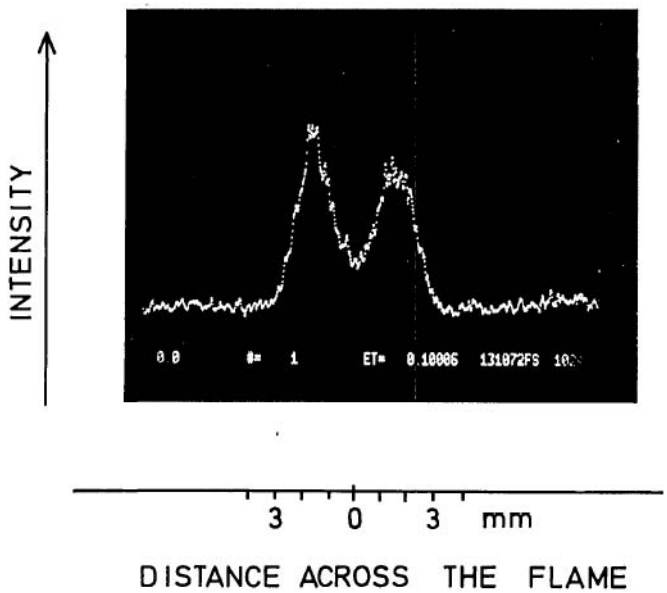

Fig. 3. Single-shot registration of the spatially resolved distribution of oxygen atoms in the lower part of a lean acetylene–oxygen flame.

the strongest transition to the $^3P_2$ state. The direct and collisionally assisted decays are illustrated in Fig. 2(a), which shows the fluorescence spectrum of O obtained in a flame using ten laser pulses, each having an energy of 1.0 mJ. For comparison a measurement with the same experimental conditions was performed in air [Fig. 2(b)] indicating no presence of oxygen atoms. Here a 500-mm quartz lens was used to focus the UV laser beam. This long focal length was chosen to minimize the influence of high-power densities and breakdown in the focal region and subsequent creation of oxygen atoms by photodissociation and other processes. With this lens no detectable influence of the laser beam was observed. Using lenses with shorter focal lengths ($f < 200$ mm), oxygen atoms were created by the intense laser beam producing a spurious oxygen atom signal.

The total fluorescence intensity was used to image the radiation from oxygen atoms on the horizontally mounted diode array. A Schott RG 715 colored glass filter provided sufficient isolation of the fluorescence light. Figure 3 shows a single-shot recording (1.0 mJ) of the oxygen atom distribution along the laser beam in the lower part of the acetylene–oxygen flame. The peak signal in a single laser pulse was of the order of 600 counts/pulse, which occurred in the reaction zone. This zone in an atmospheric pressure acetylene–oxygen flame is very thin (<50 μm). Since the two-photon excitation process exhibits a quadratic laser intensity dependence, the shape of the beam waist is of importance in imaging experiments. However, with the long focal length lens used above no need for correction due to this effect was found.

As demonstrated in this Letter laser-induced fluorescence can be used for spatially resolved detection of oxygen atoms. Thus the technique has in addition to its nonintrusive feature a distinct advantage over the optogalvanic technique, where only point measurements are feasible. A potential problem here as well as in other techniques using high power laser beams is the creation of species by the focused laser beam. As already mentioned, it was realized in our experiments that oxygen atoms were created by the laser beam when using a too short focal-length lens. This has also been observed by Miziolek.[10] Thus great care has to be taken to make correct measurements.

In this work no attempts were made to estimate the oxygen atom concentration from the strength of the signal. However, in a previous study on a similar flame,[5] such calculations were performed yielding an oxygen atom number density which was ~2 orders of magnitude lower than the expected value (5–10% mole fraction). This large discrepancy between theory and experiment has also been experienced by Bischel *et al.*[11]

We plan to use this imaging technique in the near future for spatially resolved detection of O-atoms as well as OH and CO moelcules in ignition experiments to follow the dynamics of these processes.

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Linearity study of a diode array radiometer

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In the course of applying a silicon photodiode array spectroradiometer to recent measurements of light sources, we have studied the linearity of response of our instrument. This study indicates that our instrument is capable of radiometric measurements to an accuracy of ~1% without applying corrections for departure from linearity. This is a significant improvement over an earlier model which we examined. The silicon diode array detector in this radiometer consists of a linear array of 1024 discrete photodiodes which are reverse biased so that they behave effectively as charged capacitors. As light strikes a diode, electrical charge carriers are generated which tend to discharge this capacitance. The electronic controller, meanwhile, periodically accesses each diode and recharges it. The measurement of the charge required in recharging is digitized and displayed by the radiometer as a measure of the accumulated radiant energy incident on that diode since its last previous recharge. The diode array is mounted in the focal plane of a small grating spectrograph so that each diode corresponds to a wavelength bandwidth of ~0.5 nm.[1]

For determination of the response function we employed an automated beam conjoiner.[2] In this device, the flux from a constant source is split into two branches by a beam splitter. The flux in each branch is attenuated by one of four filters mounted on a filter wheel or blocked by an opaque portion of the wheel. The two fluxes are then recombined by a second beam splitter and further attenuated by one of four filters on a third wheel. The available combinations of the twelve filters allow for 96 levels of flux, ranging over a factor of ~500. A microcomputer controls the stepping motors which position the filter wheels and records both the filter wheel positions and the radiometer response for each measurement. An experiment consists of the 96 measurements of flux, presented in random order, interspersed with 29 dark response measurements. Since the flux contribution from both branches when measured together is equal to the sum of the fluxes from each branch when measured separately (additivity), such an experiment permits the relative values of flux to be derived and compared to the radiometer response signal, provided one can assume a simple functional relationship between flux and signal.[2,3]

In the experiments to be reported here we examined only the dependence upon incident radiant energy of the relative response function (i.e., linearity) of the radiometer system. We studied the effect of diode position by recording data from eight adjacent diodes near the center and near each end of the array. We studied the effect of different wavelengths of light upon the same diode by changing the rotation angle of the grating. And we studied the effect of the length of the signal

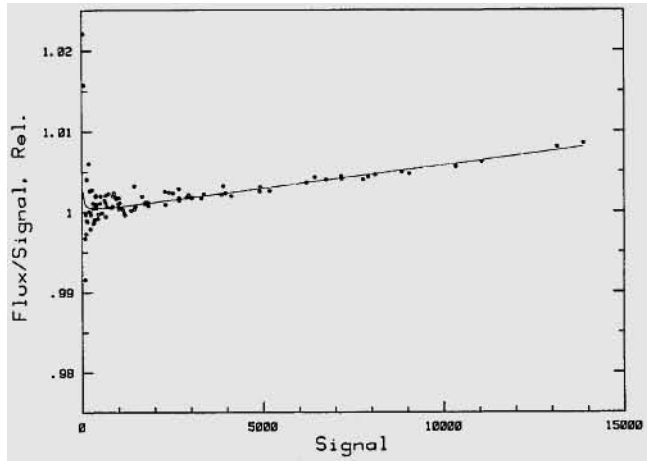

Fig. 1. Relative output signal correction factor for diode 502 irradiated at 774 nm with an accumulation time of 11.3 msec. Signal units are counts and range from 0 to 16383, where the analog-to-digital converter saturates.

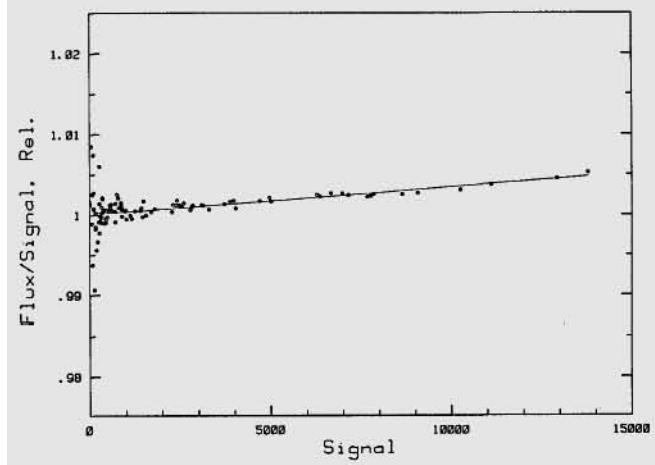

Fig. 2. Relative output signal correction factor for diode 502 irradiated at 543 nm with an accumulation time of 6.5 msec.

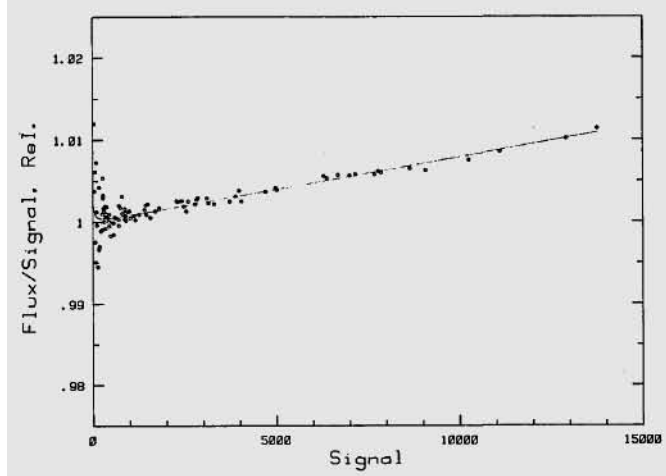

Fig. 3. Relative output signal correction factor for diode 503 irradiated at 543 nm with an accumulation time of 6.5 msec.