

Two-dimensional quenching lifetime measurement of OH: $A^2\Sigma^+(v' = 1)$ and NO: $A^2\Sigma(v' = 0)$ in atmospheric-pressure flames

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Abstract. Planar laser-induced fluorescence is currently widely applied to research on combustion. However, conventional approaches for semi-quantitative measurement could not provide the satisfactory solution and the problem due to collisional quenching remains to be solved. This paper describes the two-dimensional (2D) quenching-time-constant measurement of OH: $A^2\Sigma^+(v' = 1)$ and NO: $A^2\Sigma(v' = 0)$ in flame cross sections at atmospheric pressure. These measurements involve 1–2 ns decay time of the excited state using a high-speed image intensifier and a tunable laser with a pulse duration of 3 ns at FWHM. The correlation factors of the exponential fits for the fluorescence decays after the laser pulse were larger than 0.999 in all experiments presented in this study. Furthermore, the measured Stern-Volmer plots of the quenching rate at 1.0, 1/2, 1/3, and 1/4 atm pressure using the same experimental apparatus was confirmed to have a linear relationship for both OH and NO, showing that the 2D decay-time measurements of 1–2 ns have been successful. For NO: $A^2\Sigma(v' = 0)$, the obtained quenching rates inside the inner cone and in the outer flame in the NO-seeded methane–air Bunsen flame were 8.7×10^8 and $7.8 \times 10^8 \text{ s}^{-1}$, respectively, and for OH: $A^2\Sigma(v' = 1)$, the obtained quenching rate mapping in the outer flame was around $5.6 \times 10^8 \text{ s}^{-1}$ in the methane–air flame.

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Predissociation Fluorescence (LIPF) and the other is Laser-Induced Saturated Fluorescence (LISF). The former has been applied for OH [1,2] and CH [3], but predissociative level is often too high to be excited by the commercially available high-power lasers which are suitable for PLIF measurement. In addition, in order to obtain an effective semiquantitativeness, the molecular predissociation rate constant should be one order of magnitude greater than quenching rate constant, which causes a significant decrease in quantum efficiency of LIF measurements and this lowers the detectability of low-concentration species due to their weak signal, thus limiting the applicability of this technique. In the case of planar LISF, since an intense laser beam is necessary [4], only injection-locked excimer lasers are applicable in UV planar LISF in practice, which also limits the applicability of PLIF. In addition, problems such as the photochemical effect [5] and temperature increase during measurement due to recyclic pumping must be taken into account for the precise measurement. Clearly, a new approach to solve the problem of collisional quenching in PLIF measurement is desired.

Against this background, two-dimensional quenching variation measurement for OH: $A^2\Sigma^+(v' = 0)$ by comparing non-saturated and saturated excitation [6] by Schafer et al. has been reported. Also, there was a report of 1D decay-time measurement around 5 ns using nanosecond laser excitation which predict the 2D decay-time measurement [7], and we demonstrated 2D quenching rate measurement by means of 2D decay-time measurement for OH: $A^2\Sigma^+(v' = 0)$ in a bunsen flame cross section at half atmospheric pressure in which the collisional quenching-time constant is approximately 3 ns [8]. In our previous work [8], 2D 2–3 ns decay-time measurement was conducted using a high-speed image intensifier and a 15 ns tunable laser and 2D quenching measurement at half atmospheric pressure, which has not been successful at atmospheric pressure where many commercially available burners are used. This is due to the laser of 15 ns pulse duration which is not suitable for the high-speed decay-time measurement. In addition, the OH semi-quantitative measurement at atmospheric pressure using

Planar Laser-Induced Fluorescence (PLIF) is a two-dimensional technique to detect the fluorescence signal using an intensified camera when the target species in a laser sheet is selectively excited. Currently, PLIF is widely applied to studies on combustion; however, the error due to collisional quenching remains unsolved especially in higher-pressure measurements such as at atmospheric pressure. There have been mainly two approaches for the semi-quantitative measurement. One is Laser-Induced

molecular predissociation has been applied in many experiments and the previous report is not sufficient to show the significant potential of this 2D quenching-rate measurement by means of 2D decay-time measurement in combustion research. The experiments in this study were conducted in an attempt to solve the problem presented in our previous report.

With this objective in mind, the YAG laser with a pulse duration of 3 ns at FWHM was prepared for dye-laser pumping. In this paper, two-dimensional quenching lifetime measurements for OH: $A^2\Sigma^+(v' = 1)$ and NO: $A^2\Sigma(v' = 0)$ in the bunsen laminar flames with equivalence ratios of more than 1.0 (fuel-rich flames) at atmospheric pressure are reported because quenching lifetime distribution with a significant graduation in the outer flame was expected to be observed due to the temperature increase and the changes in species distributions due to gradual oxidation by the secondary air in the outer flame [6, 8]. For OH, the collisional quenching-time constant was observed to be approximately 10% smaller in the partially oxidized area between the inner cone and the outer cone compared with that in the outer flame, where OH fluorescence is most intense and is considered to be stoichiometrically oxidized. For NO, the collisional quenching-time constant in the outer flame has been observed to be almost flat and, at the flame front, the quenching-time constant has been observed to be slightly larger.

As to the question of how well the measurements were conducted, laser-induced fluorescence decay measurements at 1.0, 1/2, 1/3, and 1/4 atm pressure in flame cross sections were conducted using the same experimental apparatus and the Stern-Volmer plot was confirmed to be linear for both OH and NO. When the fluorescence decay signal detected by the image intensifier with a gate duration of 3 ns was exponentially fitted, the correlation factors were found to be larger than 0.999 for all decay-time measurements. Additionally, the results in this paper were compared with the conventional quenching rate measurements at low pressure [9–11] and pointwise measurements in flames using picosecond excitation [12, 13]. The obtained quenching rate for OH: $A^2\Sigma^+(v' = 1)$ in this study was almost the same as that obtained by the conventional measurements. The quenching rate for NO: $A^2\Sigma(v' = 0)$ was approximately 14–17% larger than both the calculated quenching rate inside the inner cone and the value measured by picosecond excitation in the outer flame using a methane–air flame.

1 Experimental

The experimental setup is shown in Fig. 1. The laser used was a YAG-laser-pumped dye laser. The YAG laser which was custom-made by HOYA Continuum Co. consisted of an oscillator (4 mm \varnothing rod) and two amplifiers (6 mm \varnothing rod and 7 mm \varnothing rod). The pulse duration of this laser was 3 ns (FWHM) which was generated by a short-cavity oscillator with a cavity length of 120 mm long. The dye laser used was the LPD 3000 (Lambda Physik) and its pumping beam optics were changed to those suitable for a YAG laser. The dye laser was not designed for the 3 ns pumping beam and the cavity length of the LPD 3000

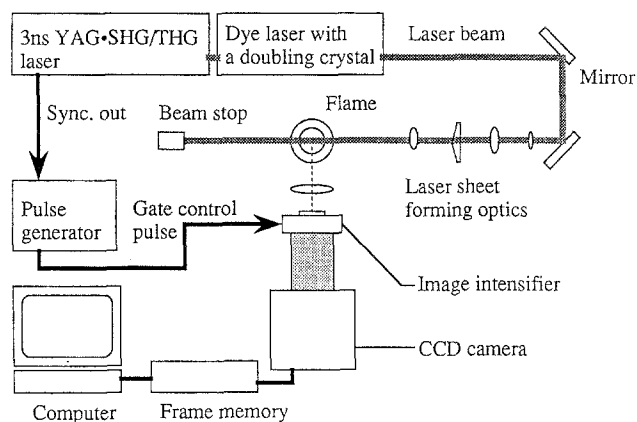


Fig. 1. Experimental setup for the 2D decay-time measurement

oscillator was approximately 300 mm long; accordingly, the bandwidth of the dye-laser output after the doubling crystal was approximately $1\text{--}2\text{ cm}^{-1}$ in our experiments. The dye-laser output was doubled by the crystal and the laser sheet was obtained after passing through a beam expander, a cylindrical lens and a prism. The laser sheet of 20 mm in height and 0.5 mm in thickness penetrated into the flame and the target species in the laser sheet plane was excited. The fluorescence signal was detected by an intensified camera positioned at 90° to the laser axis. The fluorescence image was formed on the electro-optical plane of the image intensifier through the 2:1 signal collecting optics composed of 3 lenses whose F -value was 0.75. The image intensifier used was a custom-made C4078 (Hamamatsu) which was set to 3 ns when 2D decay-time measurement was conducted. The CCD camera used was STAR 1 (Photometrics) and the CCD and the fluorescent plane of the image intensifier were 1:1 coupled by lenses. Since a single-photon signal spread to 3–5 pixels in a CCD in which a single pixel was $25\text{ }\mu\text{m}$ square, and the spatial resolution of this experimental apparatus was $150\text{--}250\text{ }\mu\text{m}$, which was confirmed by taking picture of line patterns. However, since the experiment below was conducted under the conditions of very small signal, the effective spatial resolution will be discussed separately.

In our previous work of the decay-time measurement [8], the gate of the image intensifier was optically triggered by the output laser beam to avoid jitter between the image intensifier and laser beam since the laser used was the excimer laser pumped dye laser. While, in this study, the jitter between the laser external Q -switch trigger and the laser radiation was less than 500 ps for the custom-made YAG laser. When the experiments are conducted integrating 100–300 shots of laser pulses, since the dispersion of averaged jitter is divided by the square root of number of integrated laser shots when regular probability dispersion was assumed, the expected error due to jitter would be around 30–50 ps ($500/\sqrt{100} = 300$ ps). Accordingly, the optical trigger system was not used and the gate of image intensifier and the Q -switch of the YAG laser were triggered by the same pulse generator DG535 (Stanford Research Systems).

The same burner was used for both OH and NO measurements. The inner diameter of the burner nozzle was 6 mm and the thickness of the nozzle pipe was 2 mm for flame stabilization. The flow rate of methane was fixed at 160 ml/min and the premixed air flow rate was changed with respect to the premixed equivalence ratio. For the NO measurements, 870 ppm of NO was seeded into the methane to obtain a stronger signal and to obtain the NO quenching rate both inside the inner cone and in the outer flame.

2 2D quenching measurement of OH: $A^2\Sigma(v' = 1)$

In this experiment, the excitation was $A^2\Sigma^+(v' = 1) \leftarrow X^2\Pi(v'' = 0) Q_1(7)$ absorption line of OH at 283 nm. The signal detected through a 315 ± 5 nm band-pass filter was $A^2\Sigma(v' = 1) \leftarrow X^2\Pi(v'' = 1)$ fluorescence and partially overlapping $A^2\Sigma(v' = 0) \leftarrow X^2\Pi(v'' = 0)$ fluorescence via vibrational energy transfer. The rhodamin 6G dye laser was pumped by YAG/SHG 532 nm and doubled by the KDP crystal. Figure 2a is the laser waveform at 283 nm used for the 2D decay-time measurement. The bandwidth of 283 nm laser beam was 1 cm^{-1} . Even though the pulse duration was 3 ns at FWHM, we should consider how well the 2D decay time of 1–2 ns

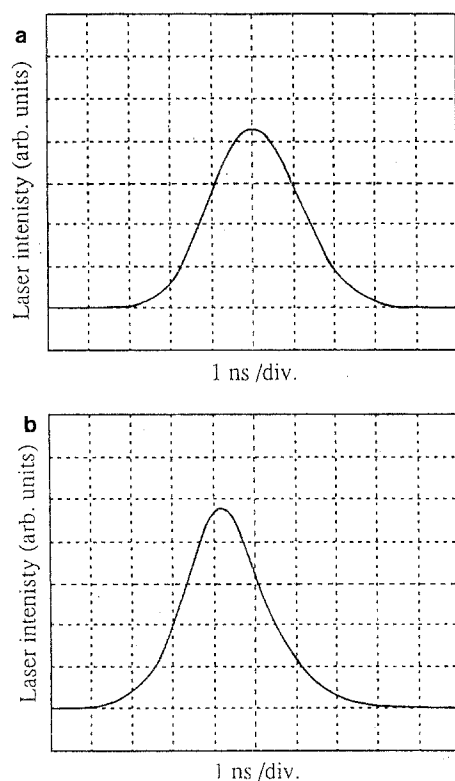


Fig. 2a,b. Typical laser waveforms measured by integrating 30 shots of laser pulses. The detector was biplanar with a rising speed of 90 ps (R1382U, Hamamatsu) and the 2 GHz-sampling digital stage oscilloscope Tektronix TDS644A. Waveform (a) is the dye-laser output at 283 nm after the doubling crystal used for OH measurements and (b) is the waveform of 226 nm output for NO measurements

could be measured. For this purpose, since the collisional quenching rate should have a linear relationship with pressure, measurements of fluorescence decay time in 1/4, 1/3, 1/2 and 1.0 atm pressure flames using the same experimental apparatus were conducted. In this experiment, the measurements were conducted at the top of the outer cone in each flame where OH concentration is high and fuel is stoichiometrically oxidized by the secondary air to avoid the dependence of the quenching-time constant on species distribution and temperature as much as possible. Figure 3 shows the laser intensity measured by the image intensifier of 3 ns gate duration using needle scattering and laser-induced fluorescence signals from the $1 \text{ mm} \times 1 \text{ mm}$ area on the laser sheet plane at 1/4, 1/3, 1/2 and 1.0 atm pressure flames with respect to 1 ns changes of the delay time. In this experiment, the fluorescence signals were integrated for every 100 shots of laser pulses. When the delay time was longer than 10 ns, fluorescence decays were observed at each pressure, and the correlation factors of the exponential fittings for the data of which delay

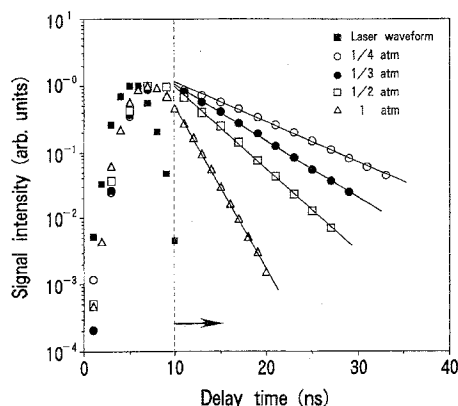


Fig. 3. Laser waveform and OH-LIF signals in outer flames at 1/4, 1/3, 1/2 and 1.0 atm measured by the image intensifier with a gate duration set to 3 ns changing the delay time

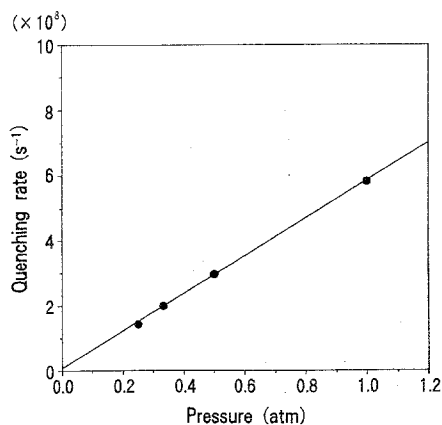


Fig. 4. The relationship between measured quenching lifetime and pressure for OH: $A^2\Sigma^+(v' = 1)$

was more than 10 ns were evaluated as larger than 0.999, indicating that the decay time was successfully measured. Furthermore, in order to confirm the reliability of the experiment, the measured quenching rate was plotted as a function of absolute pressure. Figure 4 is a Stern-Volmer plot of OH quenching rate and shows that quenching rate has a linear relationship to pressure, which means that the specifications of the experimental apparatus are suitable for the quenching measurement of OH: $A^2\Sigma^+(v' = 1)$ at atmospheric pressure. Figure 5 is an example of the laser-induced fluorescence signal obtained while changing the delay time of the image-intensifier gate every 1 ns using the flame with a premixed equivalence ratio of 1.19. The image data were acquired by integrating 300 shots of laser pulses for each image. Figure 6a shows a direct

photograph of the flame used for the measurement ($\phi = 1.19$), Fig. 6b a quenching-time-constant mapping calculated from the series of images in Fig. 5, Fig. 6c a direct photograph of the flame with a premixed equivalence ratio of 1.59, and Fig. 6d the quenching-time-constant mapping in a cross section of the flame ($\phi = 1.59$) measured by the same method as that used in the case of Fig. 6b. Figure 7a is a sample of a histogram which accumulates each pixel value in the 30×30 pixel square (1.5 mm square in the laser sheet plane) and Table 1 is the list of averaged values in 20×20 pixel squares shown in Fig. 7b. As shown in Fig. 7a, measured quenching lifetime mapping has a roughness of ± 0.1 ns, and also Table 1 shows that a reliable two-digit value can be expected with a resolution of 1 mm square on the laser sheet plane.

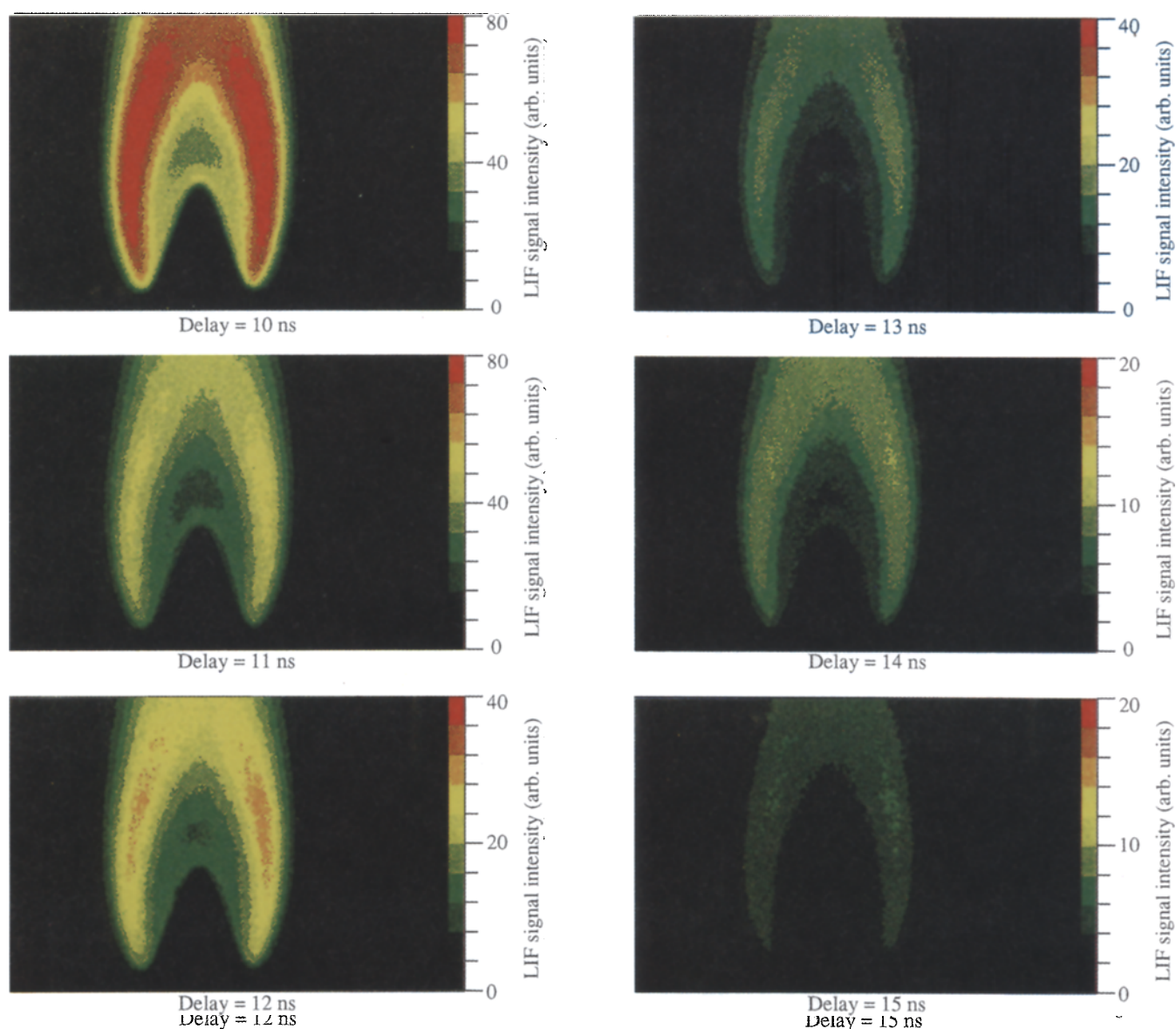


Fig. 5. An example of a series of PLIF signal decay for OH: $A^2\Sigma^+(v' = 1)$ in a cross section of a Bunsen laminar flame ($\phi = 1.19$)

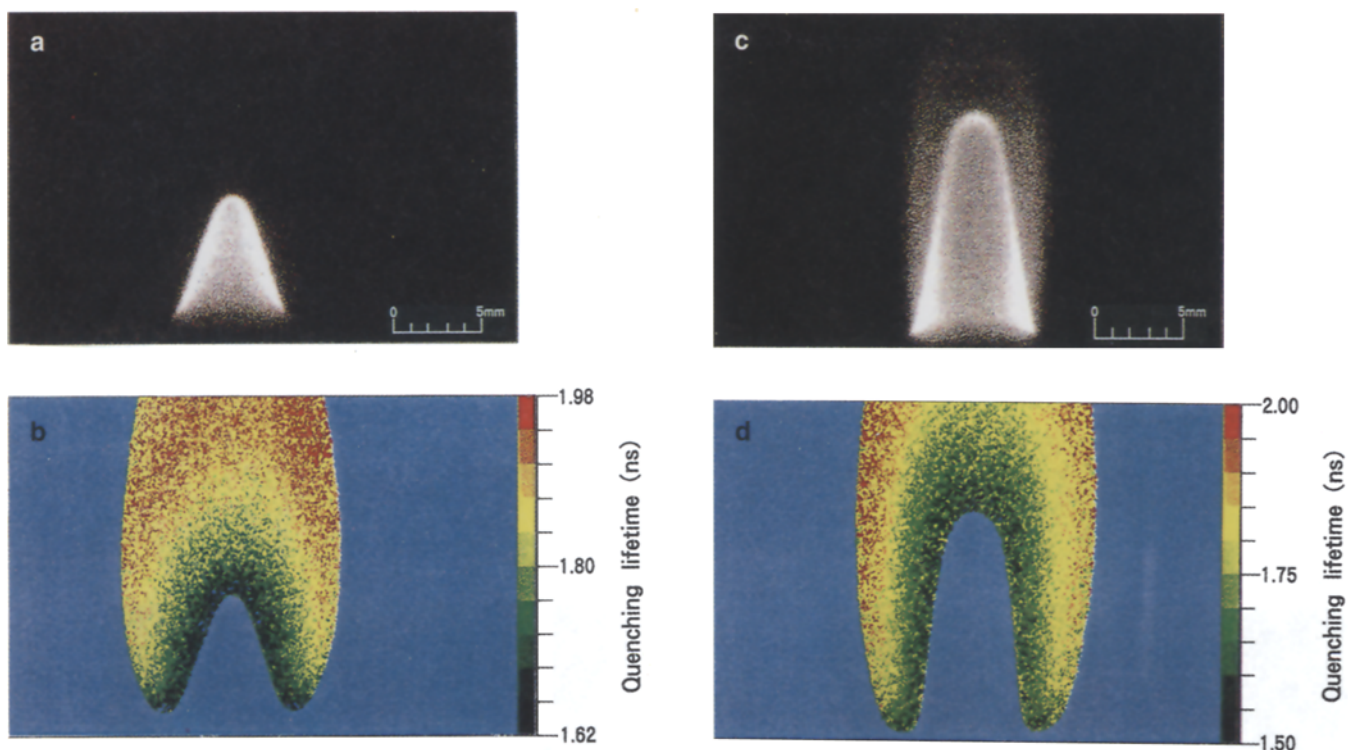


Fig. 6. **a** A direct photograph of a bunsen flame ($\varnothing = 1.19$) used for the measurement. **b** Mapping of quenching-time constant of OH: $A^2\Sigma^+(v' = 1)$ in a flame cross section of the bunsen flame ($\varnothing = 1.19$). **c** A direct photograph of a bunsen flame ($\varnothing = 1.59$) and **d** quenching-time constant mapping of OH: $A^2\Sigma^+(v' = 1)$ in a cross section of a flame ($\varnothing = 1.59$)

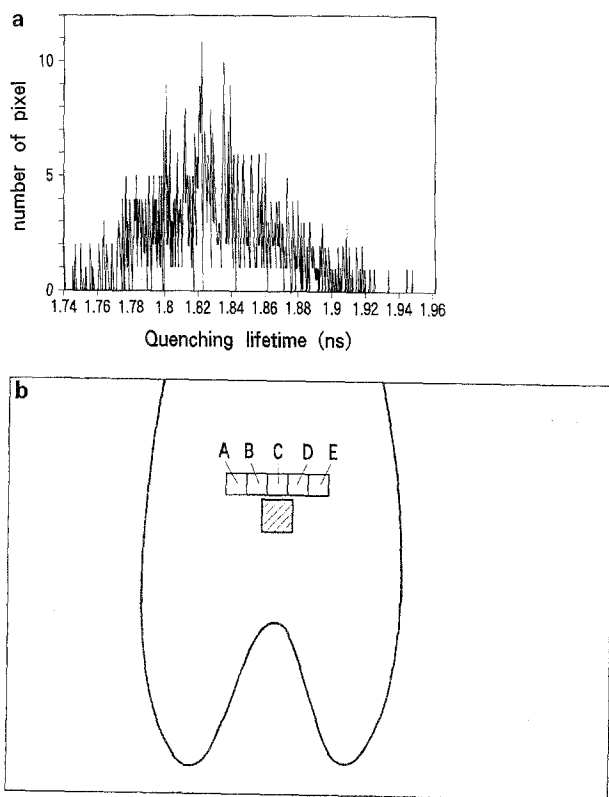


Fig. 7. **a** Distribution of pixel values in the hatched area (1.5 mm square) of the flame cross section shown in **b** which is made by tracing Fig. 6b. The list of averaged values (Table 1) in 1 mm square areas "A-E" are shown in **b**

Table 1. Averaged quenching lifetimes from Fig. 7b "A-E"

	A	B	C	D	E
Averaged quenching lifetime [ns]	1.87	1.85	1.86	1.85	1.86

3 2D quenching measurement of NO: $A^2\Sigma(v' = 0)$

In this experiment, NO: $A^2\Sigma(v' = 0) \leftarrow X^2\Pi(v'' = 0)$ $Q_1(28)$ absorption at 266 nm was excited by a YAG/THG-pumped coumarin 47 dye laser with a doubling crystal. The output of YAG/THG was 120 mJ/pulse and its pulse duration shown in Fig. 2b was approximately 3 ns. The laser output after the doubling crystal was 1 mJ/pulse with a pulse duration of 2.5 ns and bandwidth of 2.0 cm^{-1} . Since NO absorption lines lie close to each other because of the small value of the B_v molecular constant, even though the laser was tuned to the $Q_1(28)$ branch of the NO: $A^2\Sigma(v' = 0) \leftarrow X^2\Pi(v'' = 0)$ which was isolated from the neighboring lines, the laser light, of which the bandwidth was 2.0 cm^{-1} , could not selectively excite a single line perfectly and the $Q_2(31)$ and $R_2(25)$ lines were slightly excited at the same time in this experiment. To detect fluorescence signal, the band-pass filter of $248 \pm 5 \text{ nm}$ was placed in front of the image intensifier to collect the signal of NO: $A^2\Sigma(v' = 0) \leftarrow X^2\Pi(v'' = 2)$. In this

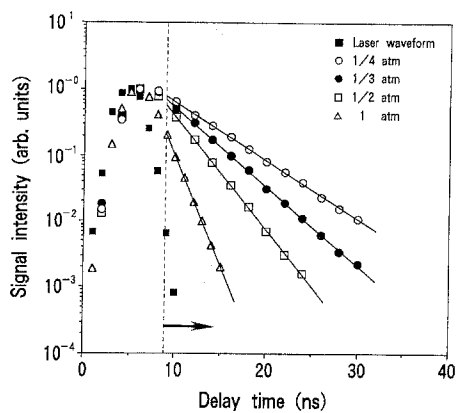


Fig. 8. Laser waveform and NO-LIF signals in outer flames at 1/4, 1/3, 1/2 and 1.0 atm pressure changing the delay time and setting the gate duration of the image intensifier at 3 ns

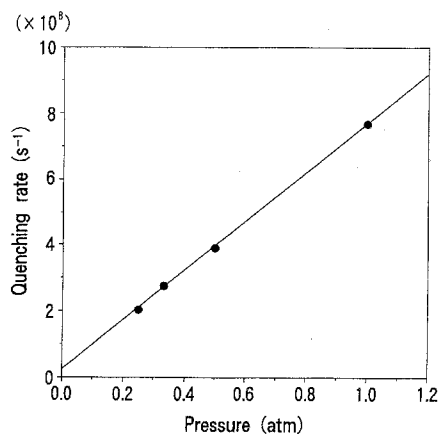


Fig. 9. The relationship between the measured quenching lifetime and the pressure for NO: $A^2\Sigma(v' = 0)$

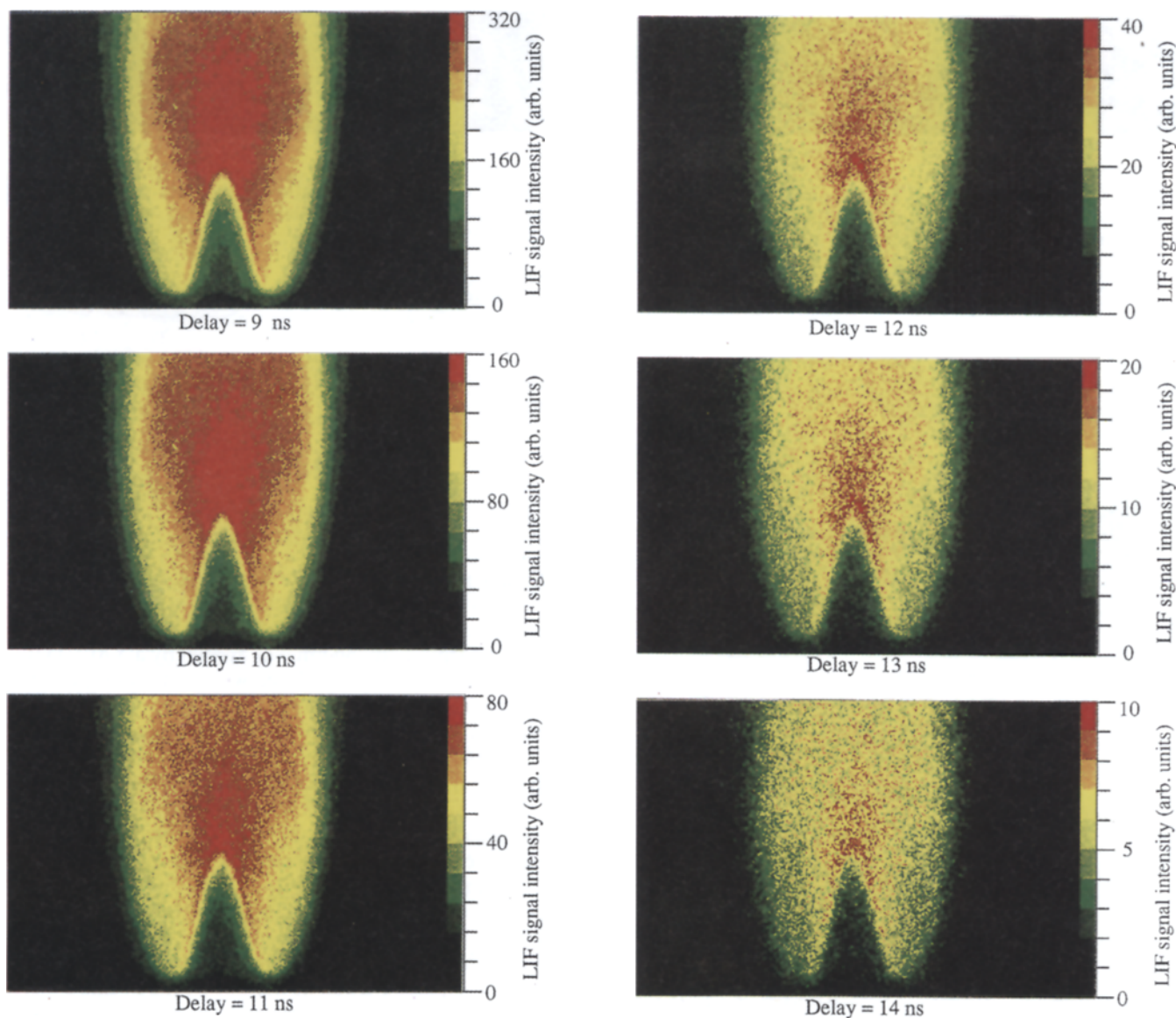


Fig. 10. An example of a series of PLIF signal decay for NO: $A^2\Sigma(v' = 0)$ in a cross section of a bunsen laminar flame ($\varnothing = 1.19$)

excitation detection scheme, significant O_2 -LIF was not observed. Regarding the PAH-LIF problem, since fluorescence decay of PAH-LIF should be shorter by more than one order of magnitude compared with NO, adverse influence upon the quenching rate measurement of NO is considered not to occur. Also, the authors confirmed that even in the normal planar laser-induced fluorescence visualization for NO using this scheme, which collects the total LIF signal by setting the gate duration of the image intensifier at around 20–30 ns, PAH-LIF is negligible compared with NO-LIF when the narrow band-pass filter (248 ± 5 nm) is attached in front of the detector.

Similarly to the case of OH measurements, measurements of the decay time due to collisional quenching have been conducted in flames at 1/4, 1/3, 1/2, and 1.0 atm. Figure 8 is the fluorescence signal and the laser waveform (needle scattering) detected by the image intensifier for which the gate duration was set to 3 ns changing the delay time every 1 ns and integrating 100 shots of laser pulses for each datum and the laser waveform (needle scattering) detected by the image intensifier for which the gate duration was set to 3 ns. The decay-time measurements were conducted at the outer cone in each flame where an intense natural luminescence was observed and the equivalence ratio including secondary air was approximately 1.0. The correlation factors of the exponential fittings for the fluorescence decays after the laser pulse (delay time ≥ 9 ns) using the total signal in 1 mm square on the laser

sheet plane were larger than 0.999, and the decay-time measurements appeared to be successful. Furthermore, a linear relationship between the quenching rate and pressure was confirmed as shown in Fig. 9, demonstrating that the measurement of the decay time of around 1.25 ns was successful using the high-speed image intensifier and the 3 ns laser.

Figure 10 shows a series of laser-induced fluorescence signals of NO obtained by integrating 300 shots of laser pulses and by changing the delay time every 1 ns. The fuel/primary air equivalence ratio of bunsen flame used was 1.19. Figure 11a shows the measured quenching lifetime mapping in a flame cross section ($\varnothing = 1.19$) using the series of fluorescence signals in Fig. 10 and Fig. 11b shows the quenching-time-constant mapping determined using the same method ($\varnothing = 1.59$). Also, it is confirmed for NO, in the same way as OH, that measured quenching lifetime mapping has a roughness of ± 0.1 ns and a reliable two-digit value can be expected with a resolution of 1 mm square on the laser sheet plane.

4 Discussion and conclusions

The conventional semi-quantitative measurement method, i.e., both the molecular predissociation and the saturated excitation, could not achieve a satisfactory solution to the problem of collisional quenching in PLIF-measurement. With this in mind, a third novel approach which is thought to be effective for PLIF measurement at atmospheric pressure has been demonstrated. This new approach is the two-dimensional decay-time measurement using a high-speed image intensifier and a tunable laser with pulse duration of around 3 ns. The tunable laser provided sufficient output energy for PLIF measurements so that both the quenching lifetime mapping and PLIF concentration imaging (the total fluorescence measurement) can be achieved without changing the setup of the experimental apparatus except for the exposure time of the image intensifier. Therefore, we believe that the system used is one of the best choices for PLIF measurement.

To demonstrate the applicability of this approach, 2D quenching measurements of OH: $A^2\Sigma^+(v' = 1)$ and NO: $A^2\Sigma(v' = 0)$ in flame cross sections were conducted.

For OH $A^2\Sigma^+(v' = 1)$, pointwise quenching lifetime measurement using picosecond excitation which evaluated the time constant of 1.7–1.8 ns at $J' = 7$ in the outer cone of methane–air flame has been reported [12]. The quenching lifetime obtained in our study is distributed, 1.65–1.85 ns in outer flame, which agrees well with the results obtained in that report [12]. In addition, the linear relationship between measured quenching rate and pressure (the Stern-Volmer plot) was confirmed and the 2D decay-time measurement of OH: $A^2\Sigma^+(v' = 1)$ at atmospheric pressure was successful.

Although the NO molecule generates the most interest in combustion studies, conventional semi-quantitative measurement techniques of molecular predissociation and saturated pumping could not be applied for its PLIF measurement in practice. However, the NO distribution zone is wide in flames, which means that the temperature range in the significant distribution zone of NO is often

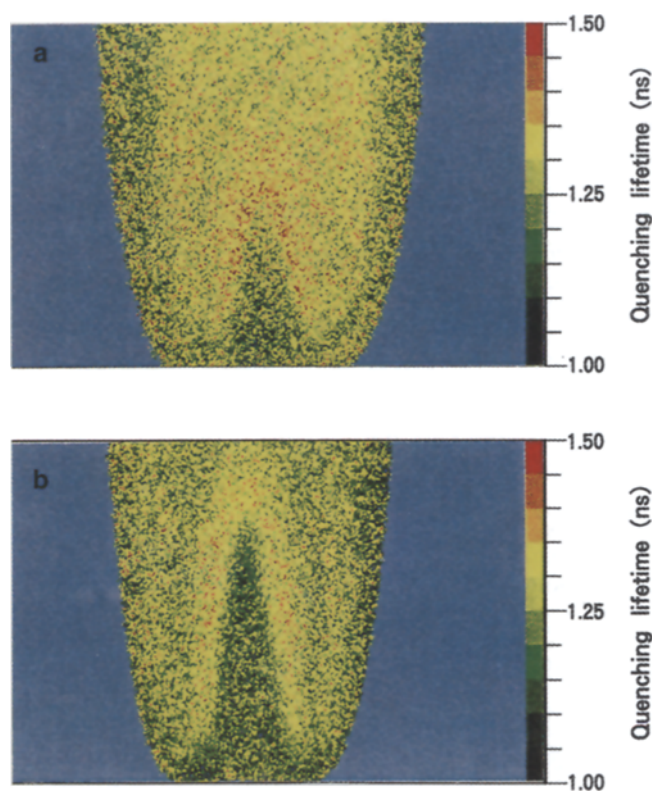


Fig. 11a,b. Measured quenching-time-constant mappings for NO: $A^2\Sigma(v' = 0)$: (a) in a flame cross section ($\varnothing = 1.19$) and (b) in a flame cross section ($\varnothing = 1.59$)

wide enough to cause a significant change in the value of the quenching time constant. Accordingly, 2D measurement of collisional quenching rate for NO is strongly desired in combustion studies. In the same manner, the reliability of the experiment was checked by changing the pressure, and the linear relationship between the quenching rate and pressure (Stern-Volmer plot) was also confirmed for NO: $A^2\Sigma(v' = 0)$. Additionally, the correlation factors of the exponential fittings for the fluorescence decays were larger than 0.999 for the measurements at different pressures.

For NO: $A^2\Sigma(v' = 0)$, there was also a report describing pointwise measurement by picosecond excitation [13], in which the decay time due to collisional quenching lifetime in outer flame was evaluated to be around 1.5 ± 0.2 ns in methane-air flames by seeding 1–2% NO when NO was excited by the laser centered at 226.4 nm for which the bandwidth was 2 cm^{-1} where the rotational numbers of P_1 head were around 7–10. On the other hand, the quenching lifetimes obtained in the outer flame in our study by seeding 870 ppm NO were about 1.29 ± 0.1 ns for $\phi = 1.59$ flame and 1.30 ± 0.1 ns for $\phi = 1.19$ flame. In addition, the quenching lifetimes inside the inner cone (unburned zone) measured in our experiment was around 1.17 ± 0.1 ns for $\phi = 1.59$ flame and 1.20 ± 0.1 ns for $\phi = 1.19$ flame. In this area, the collisional quenching is dependent on O_2 concentration because collisional quenching cross section of CH_4 and N_2 for NO molecule is negligible compared with O_2 . From the report of Paul et al. [11], the quenching cross section of O_2 for NO: A-state at 300 K is $\sigma = 25 \times 10^{-20} \text{ m}^2$, which is determined by exciting the $P_1(17)$ line. Using this value, the calculated quenching lifetime inside the inner cone at atmospheric pressure was estimated as 1.4 ns. The center value of measured quenching-time constant inside the inner cone is also smaller by approximately 14–17% than the predicted value obtained by assuming that there is a small rotational number dependence of the quenching-time constant on J' number of NO: $A^2\Sigma(v' = 0)$ [15]. The quenching-time constant obtained using the flames ($\phi = 1.19$, $\phi = 1.59$) was largest around the flame front, and was observed to be slightly smaller in the partially oxidized zone in the outer flame and smallest inside the inner cone. However, the measured quenching lifetime is distributed within $\pm 15\%$ including inside the inner cone, and, especially in the outer flame, the observed quenching lifetime was almost flat, which was in good agreement with the report of the picosecond excitation [13]. This means that the effect of the temperature range in the flame on the quenching-time constant is cancelled by the presence of H_2O and CO_2 in methane-air flames as well as OH since the collisional quenching cross sections of H_2O and CO_2 and larger than that of O_2 and their concentrations are also significantly high in the outer

flame. Accordingly, for the qualitative measurement of NO, excitation from $X^2\Pi(v'' = 0, J'' = 22\text{--}23)$ best maintains the linear relationship between the fluorescence signal and the molecular number density in PLIF measurements for methane-air flames.

To improve further this 2D decay-time measurement technique, the use of a dye laser with a shorter-cavity oscillator is recommended. In our experiment, the cavity length of the dye-laser oscillator was about 300 mm, which means that only a 1.5 round trip could be achieved with the 3 ns pumping laser. If a laser with shorter-cavity oscillator were prepared, the bandwidth of the laser would have satisfactory resolution for the single-line excitation.

In the case of the single-shot measurement for the turbulent flame application, a shorter-pulse laser with sufficient output power should be used to increase the effective signal for the decay-time measurement. This is because most of the laser-induced fluorescence was not used in order to evaluate the quenching rate in our experiment to avoid the weak excitation by the tail end of the laser pulse, and LIF signal had been integrated for 100–300 shots of laser pulses in the experiments described in this paper. Additionally, since 3 ns gate duration is not essential, we suggest that a somewhat longer gate duration of the image intensifier would also be effective in the single-shot measurements to obtain an increased effective signal.

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