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# Interference-Free Laser-Induced Fluorescence Imaging of Atomic Hydrogen in Flames

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**Abstract:** We investigate two-photon laser-induced fluorescence imaging of atomic hydrogen in flames using picosecond- and nanosecond-duration laser pulses. Picosecond excitation produces significantly larger signals for interference-free detection in methane and hydrogen/oxygen flames.

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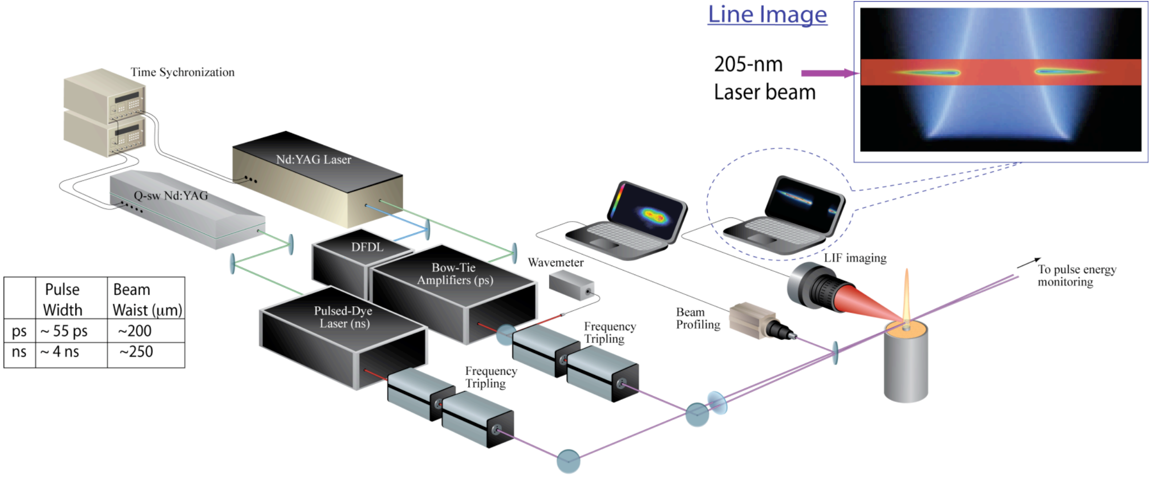
## 1. Introduction

Two-photon-resonant, laser-induced fluorescence (TP-LIF) is used widely for measuring atomic hydrogen in flames and plasmas. The simplest scheme employs two 205-nm photons for (n=1→→n=3) pumping and fluorescence detection from the $H_\alpha$ transitions (n=3→n=2) at 656 nm. In TP-LIF schemes, quantitative H-atom detection is often complicated by photolytic production of atomic hydrogen by the laser pulse [1,2] and stimulated emission (SE) [3,4]. Photochemical production of hydrogen atoms can result from photodissociation of vibrationally-excited water vapor [1] and hydrocarbon radicals, such as $CH_3$ [2]. Stimulated emission and photochemical interferences depend on the local temperature and chemical composition [5], a point that is critical in evaluating the significance of interference in two-dimensional imaging of atomic hydrogen in flames.

Because the TP-LIF signal scales as the square of the laser energy, the contribution of linear photolytic interferences can be reduced significantly by using low-energy (but high-intensity) ps excitation as opposed to commonly used ns laser excitation, as previously demonstrated for TP-LIF detection of atomic oxygen [6,7]. In the current study, we investigate TP-LIF imaging of atomic hydrogen in flames using ps and ns laser pulses.

## 2. Experimental Apparatus and Procedure

The experimental apparatus (Fig. 1) includes a tunable nanosecond laser system, a tunable picosecond laser system, an axi-symmetric premixed flame, and an intensified CCD camera for line imaging. The ns-laser system is a conventional Nd:YAG-pumped dye laser system. The picosecond laser system is an amplified distributed-feedback dye laser (DFDL) [8]. The 615-nm outputs from each laser system were frequency tripled to generate 205-nm radiation for excitation of atomic hydrogen. The ns and ps pulse durations were approximately 3.5 ns and 55 ps, respectively.



| | Pulse Width | Beam Waist (μm) |
|---|---|---|
| ps | ~ 55 ps | ~200 |
| ns | ~ 4 ns | ~250 |

Fig. 1. Experimental Apparatus

H-atom imaging was investigated in premixed $CH_4/O_2/N_2$, $H_2/O_2$, and $H_2/O_2/N_2$ flames for a wide range of flame equivalence ratios. Line images of the H-atom fluorescence (along the radial direction of the axi-symmetric laminar flames) were recorded using an intensified CCD camera. Images were collected for a range of laser pulse energies in each flame. Direct comparison of the images enabled the determination of interference-free detection limits for each flame condition. Simultaneously, SE signals were monitored in the forward-propagation direction of the laser.

## 3. Results and Discussion

For all methane and hydrogen flames investigated, LIF line images recorded for a range of ns and ps laser pulse energies are compared directly. For example, the peak-normalized LIF signals in the stoichiometic $CH_4/O_2/N_2$ and $H_2/O_2$ flames are plotted as a function of the radial position in Figs. 2 and 3, respectively.

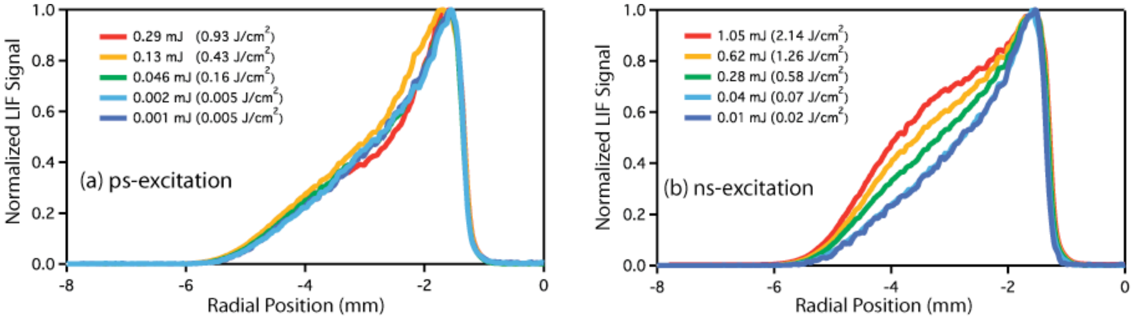


Fig. 2. Normalized TP-LIF signal in stoichiometric $CH_4/O_2/N_2$ flame using ps and ns pulses.

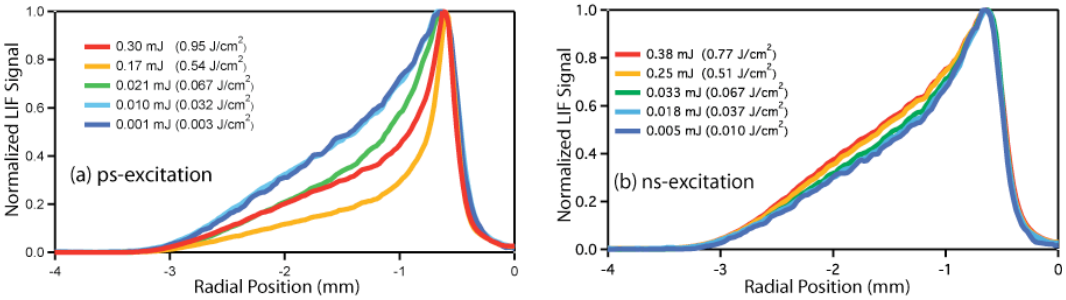


Fig. 3. Normalized TP-LIF signal in stoichiometric $H_2/O_2$ flame using ps and ns pulses.

At the lowest laser pulse energies used (blue curves), the shape of the fluorescence profiles does not change with increasing pulse energy. At higher pulse energy, however, distortions in the profile become apparent. Three types of profile distortions were observed. In the first type, a systematic change in the relative fluorescence profile occurs on the product side of the flame, as seen with ns excitation in Figs. 2 and 3. For both ns and ps excitation, this type of profile distortion is exacerbated in lean flames, for which the ratio of the number density of water vapor to that of atomic hydrogen is significantly higher than in rich flames. This observation is consistent with an interference resulting from photodissociation of $H_2O$ [3]. The second type of profile distortion is observed in stoichiometric and moderately-rich ($\Phi = 1.0 - 1.50$) methane flames with ps excitation. In this case, a change in profile shape occurs in the vicinity of the flame front, consistent with the production of atomic hydrogen via photodissociation of methyl radical ($CH_3$) in the reaction zone [4,9]. The third type of profile distortion is primarily observed with ps-excitation in stoichiometric and rich methane flames, and it is also very clearly seen in stoichiometric (Fig. 3a) and rich hydrogen flames. In this case, as the laser energy is increased, the LIF signal in the products region decreases relative to that at the peak location, resulting an effective narrowing of the relative H-atom profile. We postulate that this distortion is caused by the spatially nonuniform population loss resulting from SE. In fact, at these experimental conditions we observed strong SE signals, which could be seen easily with the naked eye.

The maximum laser fluence and peak signal level for "interference-free" imaging are listed in Tables 1 and 2 for the methane and hydrogen flames, respectively. We identify "interference-free" images as those for which there is no observable distortion in the peak-normalized H-atom LIF profile. The signal levels listed in Tables 1 and 2 correspond to the peak counts per laser shot that are collected from a volume of approximately (125-μm)$^3$.

Table 1: Peak interference-free signal levels using ns and ps excitation in $CH_4/O_2/N_2$ flames.

| | Nanosecond Excitation | | Picosecond Excitation | |
|---|---|---|---|---|
| Φ | Fluence (mJ/cm$^2$) | LIF (ave. cts./shot) | Fluence (mJ/cm$^2$) | LIF (ave. cts./shot) |
| 0.5 | 0.063 | 40 | 0.015 | 135 |
| 0.7 | 0.067 | 57 | 0.039 | 558 |
| 1.0 | 0.072 | 111 | 0.041 | 972 |
| 1.2 | 0.115 | 289 | 0.160 | 10,400 |
| 1.5 | 0.155 | 450 | 0.080 | 3,460 |
| 1.8 | 0.201 | 872 | 0.038 | 552 |

Table 2: Peak interference-free signal levels using ns and ps excitation in hydrogen flames.

| | Nanosecond Excitation | | Picosecond Excitation | |
|---|---|---|---|---|
| Φ | Fluence (mJ/cm$^2$) | LIF (ave. cts./shot) | Fluence (mJ/cm$^2$) | LIF (ave. cts./shot) |
| $H_2/O_2$ | | | | |
| 0.25 | 0.073 | 220 | 0.716 | 66,500 |
| 0.75 | 0.070 | 976 | 0.144 | 26,300 |
| 1.0 | 0.067 | 1,230 | 0.032 | 13,100 |
| 1.25 | 0.167 | 7,950 | 0.029 | 17,100 |
| $H_2/O_2/N_2$ | | | | |
| 0.62 | 0.492 | 16,200 | 0.073 | 14,900 |
| 1.0 | 0.489 | 30,400 | 0.152 | 4,460 |

## 4. Conclusions

TP-LIF imaging of atomic hydrogen using 205-nm laser pulses with ps and ns duration was investigated. The peak laser fluence for interference-free imaging of atomic hydrogen was determined by comparing peak-normalized, H-atom LIF line images. Picosecond excitation provides a significant advantage in $CH_4$ and $H_2/O_2$ flames, and ns excitation is advantageous in the relatively cooler $H_2/O_2/N_2$ flames.

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