Strategy of interference-free atomic hydrogen detection in flames using femtosecond multi-photon laser-induced fluorescence

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ABSTRACT

Hydrogen atoms are key species in combustion of hydrogen/hydrocarbon fuels. Interference-free detection of hydrogen atoms natively generated in flames using femtosecond laser-induced fluorescence (LIF) was investigated employing two colors, i.e., 243 nm and 486 nm, as excitation source: two-photon excitation followed by a relay one-photon excitation. This strategy was compared with another commonly adopted two-photon LIF strategy using 205 nm for excitation. The potential interferences were investigated, and a direct verification method was proposed to prove this strategy be interference-free, and imaging of hydrogen atoms natively generated in methane/air flames was achieved.

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Introduction

Hydrogen atoms are key species in combustion of hydrogen/hydrocarbon fuels [1], involved in flame ignition, propagation and extinction etc. Nonintrusive, spatially/temporally resolved and interference-free detection of H atoms in combustion environments will provide crucial information for fundamental understanding and practical application of combustion processes.

Various diagnostics techniques have been adopted for atomic hydrogen detection in flames, e.g., stimulated emission [2], multi-wave-mixing spectroscopy [3,4], polarization spectroscopy [5,6], laser-induced grating spectroscopy [7], and laser-induced fluorescence (LIF) [8−16], among which LIF is the most extensively used technique owing to its imaging ability with high spatial and temporal resolution, which is especially demanded in turbulent combustion research.

In traditional LIF measurement of atomic hydrogen, nanosecond (ns) laser was employed. Since the excitation of atomic hydrogen in flames can only be accessed through two- or multi-photon process [13], high pulse energy UV laser is required. The high energy UV laser, however, is powerful enough to photolyze other combustion intermediates resulting in substantial excited photolytic hydrogen atoms [17], which will generate interference to the LIF from natively generated hydrogen atoms in the flame. For instance, Kulatilaka et al. [18] reviewed this issue in detail in methane flames

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with 205-nm-laser as excitation source, and they believe that vibrationally excited H_2O, OH, CH_3 and C_2H_2 are the main precursors of photolytic H atoms. In order to suppress the photolytic interferences, researchers have utilized picosecond (ps) laser as excitation source [12—14], taking advantage of its high-peak-power but low-energy property [15]. Recently, Kulatilaka et al. [15,16,19] took femtosecond (fs) laser as an even better option for atomic hydrogen excitation due to the same reason. They adopted a strategy of two-photon excitation using a 205 nm fs laser. They developed an indirect process validating the strategy being interference-free. Excitation and detection strategies of various H-atom detection techniques are listed in Table 1.

In this work, we developed a multi-photon strategy for atomic hydrogen detection in flames using an fs laser. This strategy was compared with the two-photon LIF strategy using a 205 nm laser. A direct verification method was proposed to prove this strategy be interference-free, and imaging of hydrogen atoms natively generated in methane/air flames was achieved.

Methodology

The experiments were performed in a piloted CH_4/air jet flame burnt on a modified McKenna burner (Holthuis & Associates) which consists of a central tube (3 mm in inner diameter) and a water-cooled annular porous plug (60 mm in diameter), providing a stable Bunsen-type flame for averaging measurements. The tube and the plug can be supplied independently with premixed CH_4/air gases through separated mass flow controllers. The photo of the flame can be seen in Fig. 1 with the central jet flame on the tube being piloted by the flat flame attached on the plug. Equivalence ratio is 0.8 for the piloted flat flame, and 1.2 for the central jet flame.

The schematic of the optical setup is illustrated in Fig. 1. The laser source is a fs Ti:sapphire laser system (Spectra-Physics, Spitfire Ace) pumping an optical parametric amplifier (OPA, Light Conversion, TOPAS-Prime). The Ti:sapphire laser output has a pulse energy of 4 mJ centered at 800 nm with a pulse duration of ~45 fs and a repetition rate of 1 kHz, which was converted to a 486 nm fs laser through the OPA. The second harmonic (243 nm) of the 486 nm laser was generated
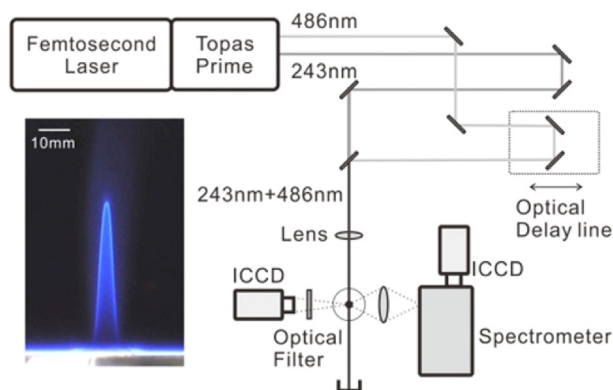

Fig. 1 — Illustration of the optical setup of fs MPLIF.

afterward in the OPA, and both laser beams were used for multi-photon atomic hydrogen excitation. The two laser beams were guided separately by various high reflection mirrors, and an optical delay line was used in the propagating path of the 486 nm beam to compensate the optical path difference of the two beams. Lastly, the two beams were combined and focused at the middle of the flame cone by a lens. A spherical lens (f = 300 mm) was adopted during spectral measurements, with which two overlapped laser beam lines were formed. During imaging measurements, it was replaced by a cylindrical lens (f = 300 mm), with which two overlapped laser beam sheets were formed. The energy density for the 243 nm fs laser at the focus is 1.6×10^3 mJ/cm^2 for spectral measurements, and 13 mJ/cm^2 for imaging measurements.

Excitation and detection strategy of atomic hydrogen using the fs multi-photon LIF is shown in Fig. 2. Atomic hydrogen is electronically excited through a two-photon process by the 243 nm laser from the ground state (n = 1) to its first excited state (n = 2), where H* is instantaneously re-excited through another one-photon process by the relay laser (486 nm) to its third excited state (n = 4). In order to avoid the stray light interference from the 486 nm laser, a non-resonant strategy was used for fluorescence detection, i.e., the secondary fluorescence at 656 nm (n = 3 → n = 2) was collected instead of the resonant fluorescence at 486 nm (n = 4 → n = 2).

Table 1 — Excitation and detection strategies of various H-atom detection techniques.

Method	Excitation		Detection		Reference
	Transition	Wavelength (nm)	Transition	Wavelength (nm)	
SE	n = 1 → n = 3	2 × 205	n = 3 → n = 2	656	[2]
4WM	n = 1 → n = 2	2 × 243	Scattered beam		[3]
6WM	n = 1 → n = 2, n = 2 → n = 3	2 × 243 + 656	n = 3 → n = 2	656	[4]
PS	n = 1 → n = 2, n = 2 → n = 4	2 × 243 + 486	n = 4 → n = 2	486	[5,6]
LIGS	n = 1 → n = 2, n = 2 → n = 3	2 × 243 + 656	n = 3 → n = 2	656	[7]
	or	or	or	or	
	n = 1 → n = 2, n = 2 → n = 4	2 × 243 + 486	n = 4 → n = 2	486	
LIF	n = 1 → n = 2, n = 2 → n = 3	2 × 243 + 656	n = 3 → n = 2	656	[10]
	n = 1 → n = 2, n = 2 → n = 4	2 × 243 + 486	n = 4 → n = 2	486	[11]
	n = 1 → n = 4	3 × 292	n = 4 → n = 2	486	[9,12]
	n = 1 → n = 3	2 × 205	n = 3 → n = 2	656	[8,13—16,19]