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Single-shot, spatially-resolved stand-off detection of atomic hydrogen via backward lasing in flames

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Abstract

We report on an experimental demonstration of spatially-resolved detection of atomic hydrogen in flames using a single-ended configuration yielding 656-nm lasing in the backward direction upon 2-photon pumping with 205-nm femtosecond laser pulses. Spatial resolution is achieved by temporally-resolved detection of the backward lasing using a streak camera. The method is demonstrated in CH_4/O_2 flames; both in a setup consisting of two flames, with variable spacing between the flames, and in a single flame. Results from the two-flame experiment show that the backward lasing technique is able to determine changes in the separation between the flames as this distance was altered. By maximizing the temporal resolution of the streak camera, obtaining a highest spatial resolution of 1.65 mm, it is possible to resolve the hydrogen signal present in the two reaction zones in the single flame, where the separation between the reaction zones is ∼2 mm. The lasing signal is strong enough to allow single-shot measurements. Results obtained by backward lasing are compared with 2-photon planar laser-induced fluorescence (LIF) images recorded with detection perpendicular to the laser beam path and the results from the two methods qualitatively agree. Although further studies are needed in order to extract quantitative hydrogen concentrations, the present results indicate great potential for spatially resolved single-ended measurements, which would constitute a very valuable asset for combustion diagnostics in intractable geometries with limited optical access. It appears feasible to extend the technique to detection of any species for which resonant two-photon-excited lasing effect has been observed, such as O, N, C, CO and NH_3.

Keywords: Backward lasing technique; Combustion diagnostics; Hydrogen atom; Ultrashort nonlinear optics; Multiphoton processes

1. Introduction

Spatially- and temporally-resolved species concentration measurements provide key insight into the understanding of physical and chemical processes in combustion. Laser-based non-intrusive

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measurement techniques, for example LIF, have been widely implemented for combustion diagnostics. Conventionally, measurements are conducted in a right-angle setup, i.e. with a detection direction perpendicular to the path of the laser beam, requiring two optical ports. However, single-ended detection techniques are required in devices limited to only one optical access, and therefore a backward detection scheme has been investigated [1,2].

As a single-ended detection technique, primarily for atmospheric studies, light detection and ranging (LIDAR) is a well-established method for remote range-resolved measurements. Basically, a laser pulse is directed towards the region of interest, where it interacts with the medium under investigation. By detecting the backscattered radiation temporally resolved, range-resolved information about the properties of the medium transected by the laser pulse can be extracted. However, the detection sensitivity and precision of LIDAR is fundamentally limited by the non-directional nature of the scattered radiation, which is emitted more or less isotropically.

For significantly more sensitive detection, a coherent signal in the backward direction, such as in the concept of backward air lasing [3–10], would be ideal. In one backward air-lasing configuration, an extremely intense pulsed (ps/fs) laser beam is self-focused in the atmospheric air to create a filamentary medium of population inversion in air constituents, including mainly oxygen and nitrogen, which gives rise to bi-directional lasing pulses. As the backward-directed lasing pulse interacts with molecules along the beam, the signal detected at the receiver carries information about the interacting molecules. Somewhat simplified, the range resolution is directly proportional to the duration of the backward lasing pulse according to $\delta R = c \times \delta t/2$, where δR is the range resolution, c the speed of light, and δt the duration of the backward lasing pulse. With the requirement of spatial resolution in the millimeter range, which is typically needed in combustion research, the duration of the backward lasing pulse needs to be on the order of a few picoseconds. To the extent of our knowledge, the shortest backward lasing pulse reported is 85 ps, and was a 337.1-nm lasing pulse of neutral nitrogen molecules, generated from femtosecond laser filamentation in pure nitrogen using circularly polarized 45-fs laser pulses at 800 nm [9].

Besides the large-scale atmospheric remote sensing discussed above, lasing effect in some atoms and small molecules of significant importance in combustion, such as O [11], H [12], C [13], N [14], CO [15] and NH_3 [16], has been observed and analyzed, and the potential for small-scale combustion research is discussed in [15]. For example, the lasing signal was used to measure vertical profiles of species in a flat flame [11,12]. As a promising diagnostic technique, the coherent nature of lasing emission provides major advantages compared to incoherent techniques, especially the high degree of signal directionality and high signal strength. The small divergence of lasing emission is a great advantage when probing flames since the interfering emissions from the flame itself as well as laser-induced, can be reduced to negligible levels using properly placed apertures or dispersive optical elements. However, lasing techniques also possess some difficulties in terms of practical diagnostics. Very poor spatial resolution was mostly encountered, mainly because of the generation of the lasing pulse along the pump laser beam and the long duration of the lasing pulse. In addition, photochemical production of species that contribute to the lasing signal was found to be a major interference in both LIF and lasing measurements [12]. Furthermore, the lasing signal strength is not proportional to the number of excited atoms/molecules along its path until it reaches the saturation regime, where the lasing signal exhibits a linear growth [17].

Regarding photochemical interferences, it has been found in two-photon LIF that the influence of such interferences can be virtually eliminated using excitation with femtosecond laser pulses. The reason for this is that femtosecond pulses provide very high peak power, promoting efficient excitation, even with low pulse energy, which minimizes photochemical effects [18], which directly applies also to lasing generation. The use of femtosecond laser pulses also significantly reduces the impact of collisional quenching due to its ultrafast excitation as well as the picosecond-scale duration of the generated lasing pulses. Furthermore, due to the high peak power of femtosecond laser pulses, much lower pulse energy is required to reach the saturation regime. All these features facilitate signal quantification and could enable extraction of absolute species concentrations with the lasing technique.

Recently, our group obtained backward lasing from hydrogen atoms in a flame based on 2-photon deep-UV excitation and a spatial resolution of a few millimeters was demonstrated [19]. The paper is focused on the generation of lasing and fundamental spatial and temporal characteristics of the backward lasing signal. Those measurements were carried out to capture the occurrence of hydrogen atoms, which is an active radical in plasma as well as combustion chemistry, and thus of essential importance in plasma-assisted ignition/combustion [20] and lean-combustion concepts with hydrogen addition [21]. In those experiments, we used 125-fs laser pulses of 205-nm wavelength to generate 656-nm lasing pulses from atomic hydrogen in the backward direction in a premixed CH_4/air flame. The backward lasing beam showed a well-localized donut-shaped spatial profile with a small divergence of $\sim$17 mrad, and can readily be collimated for guidance towards the detector. Its pulse duration, measured with a streak camera, was found

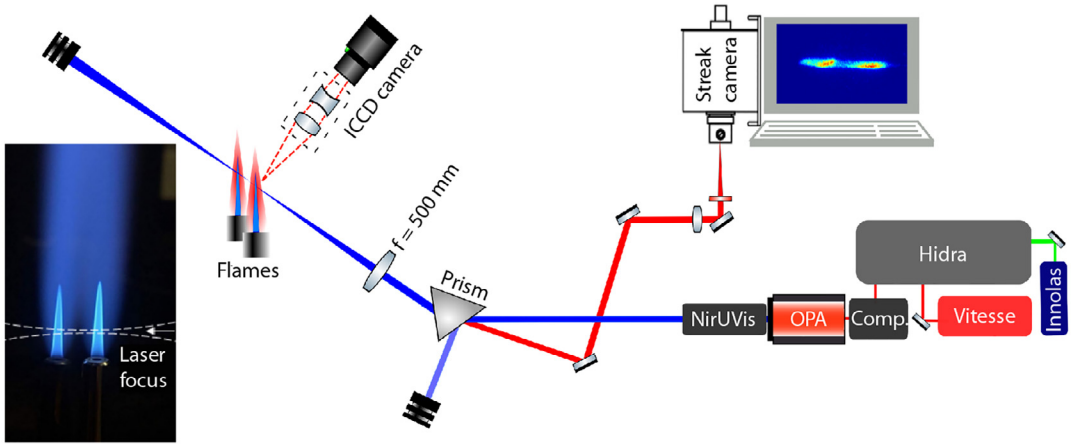

Fig. 1. Schematic illustration of the experimental setup. The inset shows luminous image of two CH_4/O_2 flames ($\phi = 1$).

to be ~15 ps. The very short time duration of the backward lasing signal constitute a major advantage compared to laser-induced fluorescence, as the signal becomes relatively immune to collisional quenching, which facilitates quantification. From two premixed CH_4/O_2 flames, burning on two small flames, two temporally-separated backward lasing pulses were generated, and a best spatial resolution of approximately 7 mm, which is actually the physical limitation of the distance between the two flames, was achieved.

Following our previous work, here we report on a thorough experimental study of this novel diagnostic technique. In experiments with two CH_4/O_2 flames, it is found that the separation between the hydrogen lasing signals, generated by the two flames, is linearly dependent on the actual separation between the flames with a constant of proportionality close to unity, proving that the measurement concept provides reliable spatial resolution along the pump laser beam. Furthermore, by increasing the 205-nm pump pulse energy and the temporal resolution of the streak camera, it is possible to resolve the hydrogen signal present in the two reaction zones in a single CH_4/O_2 flame, and a best spatial resolution of 1.65 mm is achieved. The hydrogen atom detection limit in the present experiment is estimated to ~500 ppm.

2. Experimental setup

The experimental setup is schematically illustrated in Fig. 1. A Ti:Sapphire Chirped Pulsed Amplification (CPA) laser system (Coherent Ltd.) operates at a repetition rate of 10 Hz, and provides 125-fs laser pulses at 800-nm wavelength with a maximum pulse energy of 30 mJ. The 800-nm laser pulses pump an Optical Parametric Amplifier (OPA) followed by a frequency mixing apparatus (NirUVis unit). The wavelength of the output beam can be tuned from 190 to 1600 nm. At 205-nm wavelength, the laser pulses have a maximum energy of approximately 55 μJ, and the beam diameter is about 5 mm.

The 205 nm laser beam is firstly sent to propagate through a bulk CaF_2 equilateral dispersive prism with an incident angle of 31.6°, in order to spatially separate the backward-propagating 656-nm lasing beam from the 205-nm pump laser beam. This configuration also spectrally purifies the pump laser beam by dispersing residual frequencies from the OPA and wave-mixing apparatus. A reflection from the incident surface of the prism was used to monitor the pump laser energy. The pump laser beam is then focused by a spherical lens (f = 500 mm) midway between two CH_4/O_2 flames (see the inset in Fig. 1), and creates a two-photon excitation volume of ~100 μm diameter. The laser pulse energy in the probe volume is reduced to approximately 30 μJ where most of the losses are introduced through reflections on the prism. Two welding torches are used as burner nozzles to provide narrow and stable flames for demonstration purposes. The length of the excitation volume is about 2.0 mm, varying with height above the nozzles. The diameters of the nozzles are 1.5 and 1.75 mm, respectively. In experiments with two flames, the focus of the 205-nm laser pulse is located midway between the two flames.

To estimate the separation angle between the backward 656-nm lasing beam and the 205-nm pump beam outside the dispersive prism, a He-Ne laser was employed. A streak camera (Opto-Scope S20, Optronis) with a maximum streak rate of 10 ps/mm was used to measure the temporal profile of the backward lasing pulse. The streak camera offers a temporal resolution of 2 picosecond (ps)

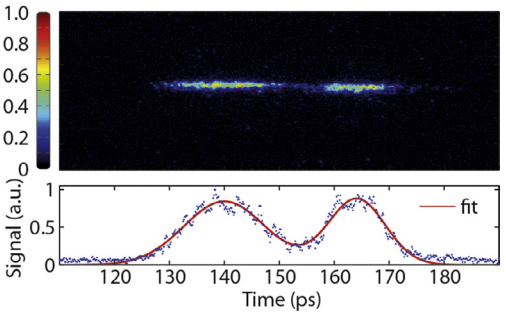

Fig. 2. Single-shot data showing the temporal profile of backward 656 nm lasing pulses originating from two CH_4/O_2 flames that are separated by 7.15 mm. The red solid line curve represents a Gaussian function fit. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

when it is operated with the fastest streak rate and a slit width of about 50 μm. A bandpass filter with a center wavelength of 656 nm (Semrock, ∼15 nm bandwidth) was put in front of the slit to improve the signal-to-noise ratio. From the side, in order to acknowledge the distance between the two volumes of H atoms, LIF images of atomic hydrogen were captured using an intensified CCD camera (Princeton Instrument, PI-MAX IV) equipped with a +50-mm-focal-length *f*/1.2 camera lens (Nikon Nikkor) and a bandpass filter centered at 656 nm.

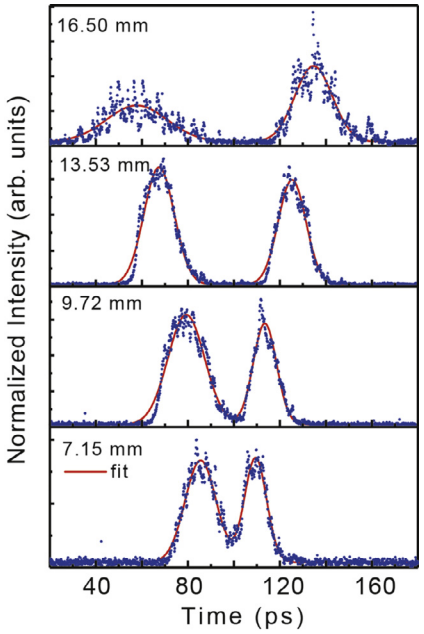

Fig. 3. Single-shot temporal profiles of backward 656-nm lasing pulses for four different distances between two flames. The red solid line curves represent fits based on Gaussian functions. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

3. Results and discussion

A single-shot temporal profile of two backward-propagating lasing signals is shown in Fig. 2. The signal originates from hydrogen atoms, which are present in the reaction zones of the two CH_4/O_2 flames ($\phi = 1$). The spatial separation between the centers of the two flames was determined to 7.15 mm from images of the flame chemiluminescence. In each of the flames, the lasing emission at 656 nm, both in the backward and forward direction, is generated by the 205-nm pump laser beam. A Gaussian function is fitted to the measured profile in order to estimate the temporal separation between the lasing pulses and the temporal duration of each pulse. The duration (FWHM) of the two lasing pulses are found to be 18 and 13 ps, respectively, and the peaks are temporally separated by 24 ps. In the backward lasing configuration, like in the LIDAR technique, the time separation between two signals corresponds to twice the distance between the two locations from which the signals were emitted. Therefore, it can be estimated that the distance between the two flames, from which backward lasing signals of hydrogen atoms were emitted, is 3.6 mm. This distance is significantly smaller than the actual distance, i.e. 7.15 mm. The discrepancy will be discussed later in the connection to Fig. 3.

Further measurements were conducted with this experimental configuration by varying the distance between the two flames while keeping the focusing of the 205-nm pump laser pulse midway between the flames. Single-shot results, obtained with the flames separated by four different distances, namely 7.1, 9.7, 13.5, and 16.5 mm, are presented in Fig. 3. One can clearly see that the temporal separations of the two backward lasing pulses become larger with increasing distance between the two flames. It is also evident that the signal strength of the lasing pulses decreases with increasing separation between the flames. This is due to the fact that the diameter of the pump beam varies with distance, having a minimum at its focus midway between the flames, and thereby the pump-beam irradiance in the flames decreases with increasing separation. It can also be noted that the peaks become narrower with decreasing separation between the flames. One possible reason for this could be that the lasing mechanism to some extent is superfluorescence, for which the duration of the lasing pulse is inversely proportional to the pump laser intensity [26].

Due to the slight difference in diameters of the two burner nozzles, it can be observed that the left peak is a little bit broader than the right one. How-

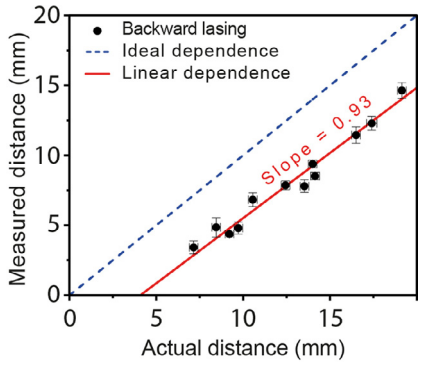

Fig. 4. The measured distance via the backward lasing technique as a function of the actual distance of two CH_4/O_2 flames. The blue dashed line is the ideal dependence whereas the red solid line represents a linear fit to the experimental data. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

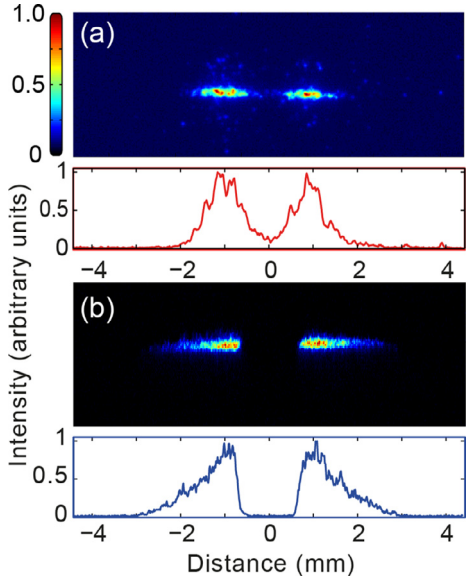

Fig. 5. (a) Single-shot measurement of the temporal profile of backward 656-nm lasing signal from single flame. The laser is focused about 3 mm above the burner nozzle. (b) Single-shot two-photon LIF image of atomic hydrogen captured with an ICCD camera.

ever, the asymmetry might be due to that the focus was not exactly in the center of the flame or exactly midway between the two flames. In principle it is possible that the lasing pulse (656 nm) generated from the second flame is attenuated when it transects the hydrogen contained in the first flame, which requires that the $n = 2$ level still contains population.

The temporal separations between the two lasing pulses were measured and converted into distances. The data points are displayed in Fig. 4. The blue dashed line represents an ideal dependence, i.e. the measured distance equals the actual distance. A linear fit to the experimental data gives a slope of 0.93 and an intercept of 4.12 mm on the horizontal axis. This linear dependence of the measured distance on the actual one suggests that the backward lasing technique can be a reliable method to spatially resolve atomic hydrogen in a flame.

The 4.12 mm intercept may be due to several factors: the non-uniform distribution of the laser intensity along the laser beam path, resulting in different delays for the generated lasing pulses, slightly different pathways of two lasing pulses and the limited temporal resolution of the streak camera.

It should, however, be pointed out that the I^2-dependence of the 2-photon absorption process has not been accounted for in these results. Such a correction, i.e. dividing by an I^2 curve, would increasingly enhance the signal with increasing distance from the focus, which would promote the signal intensity at the outer edges of the two hydrogen peaks and move the two signal peaks further away from each other, which would increase the accuracy of the measured distances and thus decrease the intercept with the horizontal axis in Fig. 4. The slit width of the streak camera was set to about 180 μm for the measurement reported above. In order to resolve the hydrogen atom concentration within a single CH_4/O_2 flame in which the peak separation of the H atoms profile is about 2 mm (corresponding to 6.7 ps), the slit width of the streak camera was minimized to 50 μm.

The concentration profile of H-atoms in the single CH_4/O_2 flame was measured through the backward lasing technique using the streak camera, and also, for comparison, by two-photon LIF with an intensified CCD camera detecting from the side.

The difference in shape between the LIF and backward lasing technique (BLT) signal profiles, as can be observed in Fig. 5, originates from the fact that for fluorescence the excited atoms interact independently, whereas for BLT the excited atoms to some extent interact collectively and the signal and its shape depend on the gain and the volume of the excited H atoms. It should be noted that the LIF profiles to some extent might be distorted by the presence of lasing since it affects the fluorescence quantum yield. However, as shown the single-shot results presented in Fig. 5, both approaches are able to resolve the hydrogen present in the two reaction zones. In this case, the actual distance between the two reaction zones was assigned to be the separation between the two peaks of the steep LIF profile. As shown in Fig. 5(b), it can be estimated to 2.0 mm. The temporal separation between the two lasing pulses, emitted from the two reaction zones separately, is 12.7 ps, which corresponds to a spatial distance of 1.9 mm, in good agreement with

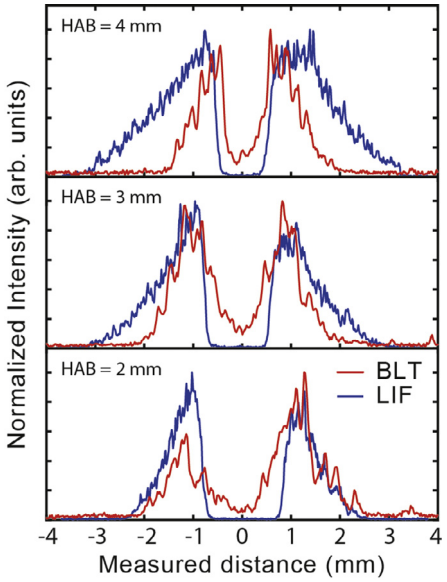

Fig. 6. Comparison between results of BLT and LIF of single CH_4/O_2 flame for different HABs. Blue line corresponds to the LIF, red line to the BLT. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

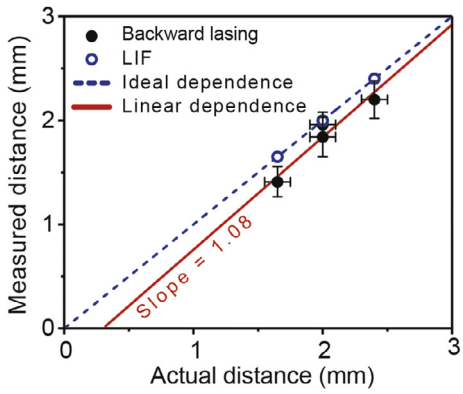

Fig. 7. Measured distance versus actual distance for backward lasing and LIF in a single CH_4/O_2 flame. The red solid line represents a linear fit to the backward lasing data, while the blue dashed line represents the ideal dependence. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

the LIF result. The temporal duration of the two pulses are 5.2 ps and 4.8 ps, respectively. Although the backward lasing technique is unable to precisely probe the steep hydrogen concentration gradient, mainly because the lasing action is a collective process with many atoms involved, it still shows the capability of millimeter-range resolution. It should be emphasized that the LIF image was obtained with the detector located only a few centimeters away from the flame, while in the backward lasing configuration an adequate spatial resolution can be achieved with a detector several meters away from the flame.

Similar experiments were further performed where the laser beam was probing at different heights above the burner (HAB), i.e. 2, 3, and 4 mm. Results from these studies are shown in Fig. 6. The same tendency can be observed for both techniques, i.e. closer to the burner nozzle surface the separation between the two peaks becomes larger. For HAB = 4 mm, the distance between the two lasing peaks, which was converted from the results obtained by the streak camera, can be estimated to 1.03 mm. This distance is the minimum distance that has been indicated with the BLT in the current experiments. However, from the LIF result, which was used as a reference, it can be seen that the two hydrogen features have maxima separated by 1.65 mm, then this distance is claimed to be the smallest resolvable separation.

Following the same procedure as for the results plotted in Fig. 4, the distance measured by backward lasing technique as a function of the actual one is shown in Fig. 7. The blue open circles correspond to LIF data and these are in excellent agreement with the ideal dependence, i.e. the blue dashed line. A linear fit (red line) to the backward-lasing data (black solid circles) gives a slope of 1.08 and an intercept of 0.3 mm on the horizontal axis. Again, the linear relation between the measured distance and the actual, now in experiments in a narrow single flame, strengthens our conclusion, i.e. the backward lasing technique is capable of resolving occurrences of hydrogen atoms.

Line-of-sight spatially-resolved detection is only possible by using the backward-propagating signal because of the single-ended configuration. A stronger backward lasing pulse with a shorter duration would improve the detection sensitivity and spatial resolution of the backward lasing technique. In a previous work [19], we have observed that the forward lasing signal is much stronger than the backward one. On the contrary, in experiments using nanosecond laser pumping, the strengths of lasing emission, following two-photon excitation, in the forward and backward direction were found to be approximately equal, however, the underlying physical mechanism is still unknown [4,15]. Therefore, a comparative study of the two-photon-excited lasing effect based on femtosecond and nanosecond laser pumping is necessary, not only for understanding the fundamental physics, but also for developing the backward lasing technique as a diagnostic.

It has been demonstrated that the use of ultrashort laser pulses can significantly reduce pho-

tolytic interferences [18,22,23]. The two-photon excitation process depends quadratically on the laser intensity, whereas the photolysis process depends linearly on the laser fluence. Thus, compared to picosecond and nanosecond excitation, a femtosecond laser provides sufficient laser intensity with significantly lower laser fluence, resulting in less photolytic interference. In our experiments, the 205-nm laser beam was focused with a spherical lens and the laser fluence at the focus (diameter of 100-μm) can be estimated to 0.32 J/cm^2. Results of radial profile measurements using LIF with picosecond laser pumping, reported in [24], exhibit some photolytic interference for two experimental conditions: (1) the laser fluence of 0.43 J/cm2 and $CH_4/O_2/N_2$ flame with ϕ =0.97; (2) the laser fluence of 0.39 J/cm^2 and $CH_4/O_2/N_2$ flame with $\phi = 1.14$. Since we used femtosecond laser pumping, it is reasonable to deduce that our measurements were free from photolytic interference. Furthermore, it was found that neither LIF nor BL was generated with the laser tuned off the two-photon resonance, which implies that no excited (at state $n = 3$) hydrogen fragments were created.

From the experimental results, we can also estimate the detection sensitivity of the single-shot backward lasing technique. Based on the following conditions: single flame ($\phi = 1$), the streak camera operating at the fastest streak rate with a slit width of 180 μm, a laser pulse energy of 50 μJ, and the laser beam focused at HAB = 3 mm, we estimate the detection limit to 500 ppm hydrogen atom concentration (with a signal-to-noise ratio of 2 as the minimum detectable signal and assuming 5% H atom concentration [25]). In order to spatially resolve the hydrogen concentration profile in a single flame, the streak camera slit width had to be reduced, with a significant reduction of lasing signal intensity as a consequence. Thus, the detection sensitivity of the backward lasing technique decreases with improved spatial resolution. With a slit width of 50 μm as applied for the result shown in Fig. 5(a), the lowest detectable hydrogen concentration is estimated to ∼2000 ppm.

4. Conclusions

Stand-off, single-shot, spatially-resolved measurements of atomic hydrogen in a single-ended configuration were experimentally demonstrated using backward lasing pulses that were generated by focusing 205-nm laser pulses of 125-fs duration into CH_4/O_2 flames (ϕ=1). The backward lasing technique was capable of resolving the occurrence of hydrogen atoms in flames separated at different distances. The linear dependence between the measured distance and the actual distance suggests a reliable method for spatially-resolved hydrogen detection. Spatially resolved H-atom distributions were captured with the backward lasing approach as well as two-photon laser-induced fluorescence in a single CH_4/O_2 flame. Rather good agreement was seen between the two measurement approaches on a single-shot basis in terms of spatial resolution, with a minimum distance resolved with the backward lasing technique in our experiments of 1.65 mm. Even though the backward lasing technique has slightly lower spatial resolution compared to LIF with 90°-detection angle, this technique can be applied for detecting atomic hydrogen in a single-ended configuration. This advantage allows the detection system to be located relatively far from the measured volume due to the collimated nature of the backward-lasing beam. The distance between the measurement volume and the detector was ∼2 m in the current investigation, but the stand-off distance could in principle be much longer since the backward-lasing signal is well collimated. Although, the current setup is capable of detecting occurrences of hydrogen atoms in flames, further investigations are needed in order to extract quantitative concentrations. This is challenging since the backward lasing signal depends nonlinearly on the number density of hydrogen, the excitation irradiance, and the temporal characteristics of the excitation pulse. Quantitative measurements rely on high detection precision, good control of the characteristics of the excitation pulse, and a valid model. Although a challenging task, quantification of the signal is possible, and work on this is already initiated in our laboratory. It should be noted though that the concept, allowing single-ended spatially resolved detection of hydrogen atoms, which has previously not been possible, provides a new tool that, already at its present status, is expected to be valuable for qualitative measurements in combustion environments. In this conceptual demonstration, where a major point was to investigate the achievable spatial resolution, it was appropriate to perform the measurements in CH_4/O_2 flames due to their relatively high hydrogen concentration (5% peak mole fraction). Nevertheless, backward 656-nm lasing from a CH_4/air jet flame has been observed [19], but the strength of the lasing was significantly weaker, due to the vastly lower hydrogen concentration (0.04%). Since resonant two-photon-excited lasing effect has been observed in O, N, C, CO and NH_3, it appears feasible to extend the present measurement concept towards single-ended spatially resolved detection of these species as well.

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