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Peculiarities of measuring fluorescence decay times by a streak camera for ps-TALIF experiments in reactive plasmas

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Abstract

In this work, the capacity of a streak camera for capturing picosecond two-photon absorption laser induced fluorescence (ps-TALIF) signals is demonstrated on decay times of H-atoms measured in low- and atmospheric-pressure plasmas. The goal is the accurate determination of the decay time of the recorded ps-TALIF signals. A ps-laser is used to excite atomic hydrogen produced in both plasmas and the fluorescence signals are detected by the streak camera using different time windows. It is shown that the choice of the time window of the streak camera affects the shapes and the decay times of the recorded raw TALIF signals. This is defined as the instrumental function of the streak camera and has a Gaussian profile as determined by recording the ultrafast laser pulse at different time windows. To remove this instrumental distortion and extract the actual TALIF signal, a dedicated mathematical procedure involving an exponentially modified Gaussian function has been developed. The application of this method leads to reliable measurements of hydrogen atoms decay times after respecting the following acquisition conditions: (i) the time window of the streak camera should be sufficiently large to capture the complete (raw) TALIF signal, and (ii) the temporal profile of the instrumental function of the streak camera should be as small as possible compared to the actual decay time of the fluorescence, while ensuring an optimal signal-to-noise ratio. This work demonstrates the remarkable potential of the combination of ps-TALIF and streak cameras in state-of-the-art optical plasma diagnostics.

Keywords: instrumental function, streak camera, ps-TALIF, collisional plasmas, hydrogen fluorescence decay time

1. Introduction

Nanosecond Two-photon Absorption Laser Induced Fluorescence (ns-TALIF) has been widely used for the characterization of various types of plasmas. Numerous studies have shown that this diagnostic can be employed to quantify atomic species in their ground states like H [1–3], O [2–4], and N [5–7]. However, in many practical cases the accurate determination of these densities via ns-TALIF becomes challenging. This happens in strongly collisional environments (from a few tens of mbar up to atmospheric or even higher pressures), where the fluorescence decay time of probed atoms is mostly on ns or even sub-ns timescales. In these cases, the use of ns-TALIF for reliable atomic density measurements can be a limiting factor as the duration of the laser pulse becomes comparable or higher than the decay time of the fluorescing atom. Thus, an ultrafast laser (e.g., using picosecond (ps) or femtosecond (fs) pulse duration) is a better choice. In fact, ultrafast TALIF avoids the need of sophisticated chemical kinetic models which are often adopted in ns-TALIF post-treatment and can bring more inaccuracies to the absolute atomic density measurements [1,8]. Therefore, accurate reactive species quenching rates can only be directly measured from their fluorescence decay signals when the laser pulse is much faster than the TALIF decay.

In addition, the use of an ultrafast detector whose response time is much smaller than the decay time of the signal is essential to effectively resolve fluorescence signal decays in ns and sub-ns timescales. In this context, the exceptional temporal resolution (Δt) of streak cameras ($\Delta t < 1$ ps [9]) is a distinct advantage in comparison to PMTs (photomultiplier tubes; $\Delta t \sim 1$ ns) or ICCD cameras (intensified charge-coupled device, minimal; $\Delta t \sim 200$ ps). Jonsson *et al.* employed a streak camera to capture ps-Laser Induced Fluorescence (ps-LIF) signals from laser-excited OH and CO in flames [10]. The use of the streak camera allowed for the measurement of the short decay time of both species reaching down to nanosecond (OH) and few hundreds ps (CO) timescales. Besides, streak cameras have been used as detectors to capture fast (ns) and ultrafast (ps) light events in plasmas. For instance,

Höft *et al.* [11] employed a streak camera (0.5 ns time window with $\Delta t \approx 5$ ps) to capture the temporal dynamics of a sub-ns pulsed spark discharge during the initial stages of its development. They also investigated the axial and radial development of a positive streamer in a dielectric barrier discharge revealing a correlation of the radial expansion with the axial propagation of the streamer [12]. Furthermore, Patel *et al.* studied an atmospheric-pressure ns-pulsed plasma by means of a streak camera using time windows between 5 and 100 ns. In this way, they were able to record the fast propagation of the discharge in the gap between the electrodes and distinguish two phases developed after the initiation of the applied voltage, i.e., a first phase between 0 and 15 ns and a second one between 15 and 100 ns [13].

However, the implementation of a streak camera in advanced optical diagnostics and the proper processing of the recorded signals is not straightforward [14,15]. Particularly, the definition of an optimal time window (hereinafter called “time range”) which is sufficient to acquire the full duration of a luminous event, depends on the characteristic timescale of the signal and the peculiarities of the streak camera. Brockhinke *et al.* applied ps-LIF in an Air/N₂/H₂ flame and measured with a streak camera fluorescence signals from laser-excited OH and H [15]. To capture the full LIF signals they used two different time windows in the streak camera: 10 ns (OH) and 0.5 ns (H). The consideration of the streak camera as detector was essential for resolving decay times as low as a few ns for OH and a few tens of ps for H. Even if the selected time range in the streak camera is the lowest offered, a signal distortion may still be present. Consequently, depending on the type of measurement performed, particular attention should be paid to extracting the actual signal by removing the artificial instrumental function of the streak camera specific to each time range. Furthermore, the selection of the slit width of the streak is crucial since it also affects the temporal resolution of the detector.

In this study, we employed a ps-laser and streak camera system to determine the fluorescence decay time of laser-excited atomic hydrogen generated in two representative types of low- and high-collisional quenching media, i.e., a low-pressure microwave discharge and an atmospheric-pressure plasma jet respectively. Application of ps-TALIF for laser-excited H-atom decay time determination is particularly beneficial owing to the short decay times of the fluorescing state in various plasma conditions. Our goals are: (i) to identify the proper time ranges of the streak camera that allow capturing different TALIF signals in each plasma, (ii) to determine and remove the effect of the corresponding instrumental function of the streak on the TALIF signal properties, and (iii) to demonstrate that a streak camera can be a suitable detector for ps-TALIF measurements in reactive plasmas. The experimental setup of ps-TALIF, plasma source and streak camera is described in section 2. Section 3 is dedicated to the presentation and discussion of the results obtained on laser-excited H-atom TALIF decay time measurements in both plasmas. The main conclusions are given in section 4.

2. Experimental setup

2.1. ps-TALIF and streak camera system

The principle of TALIF is discussed in reference [16]. Therefore, only a concise description is provided here. Briefly, atoms in their ground state are excited following the absorption of two identical UV laser photons. Then, the laser-excited atoms release their energy by isotropically emitting spontaneous photons (fluorescence). TALIF is particularly suited for atomic density measurements in plasmas or flames.

In practice, TALIF measurements of reactive atom densities are calibrated by performing the same measurements in noble gasses (such as krypton and xenon) of a known density. Thus, the fluorescence intensity can be related to the absolute density of the species being probed. In the case of atomic hydrogen, the noble gas used for calibration is krypton (Kr). The equation relating the absolute density of atomic hydrogen to the TALIF signals of H and Kr atoms is the following [1,2,4–7]:

$$n_H = n_{Kr} \frac{\int_t \int_\lambda S_{FH} \eta_{Kr} T_{Kr} \sigma_{Kr} A_{Kr} \tau_{Kr} E_{Kr}^2 \lambda_{Kr}^2}{\int_t \int_\lambda S_{FKr} \eta_H T_H \sigma_H A_H \tau_H E_H^2 \lambda_H^2} \quad (1)$$

Where n_X is the density of a species X (X here refers to H or Kr), $\int_t \int_\lambda S_{FX}$ is the temporally- and spectrally-integrated fluorescence signal of X , η_X is the quantum efficiency of the detector at the fluorescence wavelength of X , T_X is the transmission of the optics at the fluorescence wavelength of X , σ_X is the two-photon excitation cross section of X , A_X is the Einstein coefficient of the transition from the laser-excited level to a lower state of X , τ_X is the experimentally-measured fluorescence decay time of the laser-excited state of X , E_X is the laser energy selected for probing X atom, and λ_X is the laser wavelength chosen to perform TALIF measurements at the center of the absorption line of X .

In equation (1), the quantities $\int_t \int_\lambda S_{F_X}$ and τ_X are experimentally determined from raw TALIF signals recorded by means of an appropriate laser and detector system. The choice of the laser and the detector depends on the plasma condition and is crucial for an accurate quantification of τ_X [16]. For instance, to be able to record ultrafast TALIF signals in highly collisional environments (i.e., fluorescence decay times down to the ns or even sub-ns timescales), a good practice is to combine ps-TALIF with a ps detector such as a streak camera [9]. This drastically increases the accuracy of the measurement of τ_X that is directly linked to the uncertainty on the measured atomic density. Furthermore, even though a streak camera is a straightforward solution for ultrafast TALIF diagnostics, its use to acquire a ps-TALIF signal requires considering its peculiarity in terms of instrumental function that changes with the targeted time range and entrance slit width.

To perform ps-TALIF measurements in this work, a ps laser from EKSPLO[®] was used. The laser system has been described in detail in [17] and therefore only a brief description is provided here. It is composed of three units: a Nd:YLF pump laser (PL3140), which produces a fundamental output at 1053 nm, a harmonic generator (APL2100) used for fundamental frequency doubling (527 nm) and tripling (351 nm), and a solid state optical parametric generator (PG411) generating ultrafast laser pulses (≈ 10 ps pulse duration; 60 μJ /pulse energy around 205 nm) in the spectral range 193-2300 nm with a repetition frequency of 5 Hz. The experimental setup is depicted in Figure 1 comprising the excitation unit (laser), the detection unit (streak camera; see below), and two plasma sources used alternatively (here represented as a violet sphere inside a dashed square for simplicity).

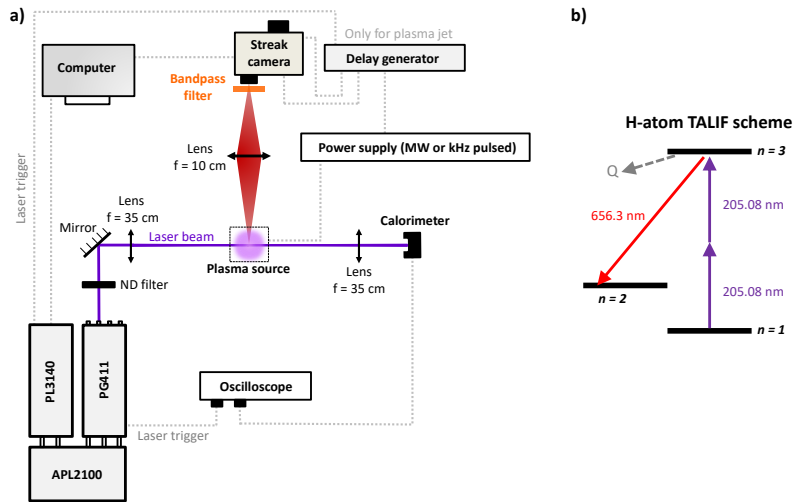


Figure 1: (a) Experimental setup of the ps-TALIF experiments performed in a low-pressure microwave plasma and an atmospheric-pressure plasma jet using a ps laser and a streak camera system. (b) H-atom TALIF scheme comprising two-photon excitation of ground state hydrogen to $n=3$, subsequent relaxation to $n=2$ through fluorescence emission at 656.3 nm (red) and collisional quenching of the laser-excited state (gray).

A diaphragm is installed at the output of the PG411 unit of the laser to obtain a circular beam profile. Neutral density filters are placed after the diaphragm to adequately tune the laser energy. In principle, a suitable laser energy (E_L) for performing H-atom TALIF should be within a regime where the TALIF signal intensity scales quadratically with E_L [16,17]. For our laser system, this is the case for $E_L \leq 14$ μJ /pulse and $E_L \leq 57$ μJ /pulse for the low- and the atmospheric-pressure plasma sources, respectively. The E_L values selected to perform H-atom TALIF experiments are 11 and 38 μJ /pulse, respectively. Furthermore, a lens with 35 mm focal length is used to obtain a laser spot with a diameter of 500 μm , which determines the spatial resolution of our TALIF system. After passing through the discharge, the laser beam is collected by another lens with 35 mm focal length and focused on a calorimeter (Coherent J-10MB-LE, 11630V/J) connected to a digital oscilloscope (Lecroy WaveSurfer 10, 1 GHz, 10 GS/s). The fluorescence is collected side-on to the laser's beam direction, avoiding a direct interaction between the laser beam and the detector. The fluorescence is imaged using an achromatic lens with 10 cm focal length. Finally, a bandpass filter (Semrock FF01-655/15-25) is placed in front of the slit of the detector to remove any stray light coming from the plasma or the surrounding environment.

The detector used is a universal streak camera (HAMAMATSU[®] C10910-05) that can capture light signals down to ps timescales. Among other components, it includes dedicated optics for light focusing on a photocathode where photoelectrons are produced, which are subsequently accelerated, swept, and multiplied by a micro-channel plate (MCP) within the streak tube to finally impact a phosphor screen [9]. There, they are converted back into light that is recorded by means of a readout CMOS camera (ORCA[®]-Flash4.0 V3 C13440-20CU). Streak cameras generally feature distinct time windows for collecting light signals in a specific time range (TR), which is called sweep time,

and has values between 100 ps and 1 ms for our streak camera system. The variation of TR is achieved by alternatively using two different sweep units: a “fast” single unit (M10912-01; sweep time: 100 ps to 50 ns; temporal resolution: <1 ps FWHM) or a “slow” single unit (M10913-11; sweep time: 1 ns to 1 ms; temporal resolution: <20 ps FWHM). The time range of the sweep units of the camera is directly connected to the steepness of the voltage ramp applied to the sweep electrodes inside the streak tube to temporally-separate the incoming photons. Thus, it is necessary to adapt the time range of the streak camera to the total duration of an incoming light event. The streak camera, the laser, and a pulsed power supply (only used when studying an atmospheric-pressure plasma jet; see below) are synchronized using a 5 Hz TTL signal (internally generated by the laser) and a couple of delay generators (Stanford Research DG645, HAMAMATSU Delay Unit C1097-05). For the fluorescence measurements performed in this study, the entrance slit width of the streak camera has been fixed at 150 μm . The streak camera is operated by a dedicated software installed on a desktop computer. All TALIF signals in this work are recorded using the following parameters: maximum MCP gain=63, number of exposures=20, and CMOS integration time=10 s, resulting in a total acquisition time (TAT) of 200 s for recording one raw TALIF signal. These are the conditions for which the best TALIF signal-to-noise ratios (SNR) were obtained in our experiments.

2.2. Plasma sources under investigation

In this work, two plasma sources are used as testbeds to record ps-TALIF signals by means of a streak camera. First, a microwave plasma reactor, which has been described elsewhere [6], is employed for performing H-atom ps-TALIF measurements at low pressures. It consists of a stainless-steel 6-way cross enclosure with a volume of about 4 L. A microwave plasma is generated in the reactor using a commercial collisional source (Sairem Hi-Wave) detailed in [18]. The microwave power is supplied by a solid-state generator (Sairem GMS 200) through a coaxial cable. The generator delivers a power of 200 W at a frequency varying between 2.4 and 2.5 GHz to obtain the best impedance matching between the source and the discharge. The cavity is pumped by a set of turbo-molecular and rotary pumps (Edwards EXT75DX and Pfeiffer Duo 6M) allowing for the gas pressure to be decreased down to 10^{-6} mbar. The pressure in the chamber is measured by a capacitive pressure gauge (Pfeiffer CCR 363) operating in the pressure range 1.33×10^{-3} –13.3 hPa. Pure hydrogen is injected by means of a flow meter (Bronkhorst EL flow) at a constant flow rate of 5 standard cubic centimeters per minute (scm). Manual control of the primary pump valve allows for the reactor’s pressure to be varied from 40 to 200 Pa (typical operating conditions for the Hi-Wave source in this work). Finally, two fused-silica windows are crossed by the laser beam, while a third one (BK7) is located at 90° with respect to it. To probe H-atoms in this type of plasma, there is no need for synchronization of the laser and streak camera system with the plasma power supply since the plasma is stationary at 2.45 GHz [6]. The interaction zone between the laser spot and the plasma is located 2 cm below the source.

Moreover, an atmospheric-pressure plasma jet is employed for H-atom ps-TALIF measurements at high pressure conditions. More details on such plasma devices and their applications can be found in reference [19]. The used plasma jet consists of a thin dielectric tube (quartz) wrapped by two ring electrodes, thus forming a dielectric-barrier-discharge-like configuration. The first ring electrode which is closer to the tube’s outlet is biased by bipolar square voltage pulses (3.5 kV peak-to-peak) with a repetition frequency of 20 kHz and a duty cycle (i.e., ratio between the pulse width and the period of the voltage) of 50%. The second electrode is directly connected to the power supply’s ground potential. The operating gas is helium (99.999% purity) at a flow rate of 1 standard liter per minute (slm) set by means of a mass flow meter (Bronkhorst MV-392-HE). The plasma source is vertically fixed on an optical table with the helium gas flowing upwards. In this configuration, the laser beam is focused just above the nozzle of the dielectric tube. This enables avoiding reflections from the glass tube that would interfere with the TALIF signal from the plasma. The plasma ignition time (t_0) can be set by the trigger of the laser, while the laser pulse starting time is adjustable with respect to t_0 using the abovementioned delay generators. All measurements are performed at the rising edge of the applied voltage pulse where an enhanced production of H-atoms by the plasma takes place.

3. Acquisition of ps-TALIF signals using a streak camera and their interpretation method

The validity of TALIF measurements of atomic densities requires a decay time of fluorescing states that is much larger than the laser pulse duration. In fact, the decay time of the laser-excited states of these atoms depends on the collisional quenching that is dictated by different factors such as the operating pressure and the nature of the dominant quencher. For instance, application of ns-TALIF for H-atom quantification in plasmas could become challenging, owing to the small natural radiative lifetime, and to the shortening of the fluorescence decay times by strong collisional quenching even at pressures as low as 1 mbar (see section 3.2). In such cases, a kinetic scheme is often used to estimate the H-atom decay times.

Thus, for conditions where collisional quenching is too strong for ns-TALIF to be applicable, as was shown in the case of N-atom produced in a micro-hollow cathode discharge ignited in pure N_2 at 60 mbar or higher pressures

[20], the consideration of ps-TALIF becomes mandatory. Indeed, ps-TALIF is particularly advantageous for probing fluorescence decay times that can fall even to sub-ns timescales. As an example, we refer to the decay time of fluorescing hydrogen atoms in a flame at atmospheric-pressure conditions [21]. Furthermore, the analysis of such very short decay times also requires the implementation of ultrafast detectors. Streak cameras are therefore the most representative among them with temporal resolutions as low as few ps (at least).

In the following subsections, we show how the features of the H-atom TALIF signal (shape, rise time, and decay time) may be affected by the peculiarities of the streak camera with emphasis being put on its time range. It must be noted that a determination of the absolute H-atom densities in both plasmas, their dependence on different plasma conditions as well the study of their kinetics is out of the scope of the present work.

3.1. Instrumental function of the streak camera

The streak camera employed in this study can capture light events in time ranges varying between 100 ps and 1 ms. This is possible by adapting the sweep time of the fast and slow sweep units. For the experiments presented here, the time range is varied between 200 ps and 500 ns and its effect on the H-atom TALIF signal properties is studied for both plasmas (i.e., TALIF signals are recorded for the same plasma conditions but using different time ranges). For simplicity, only the signals corresponding to the low-pressure plasma are illustrated in Figure 2 (spatially averaged TALIF around the center of the CMOS sensor, Figure 2(a)). These results allow for the choice of suitable time ranges that give more reliable raw TALIF signals.

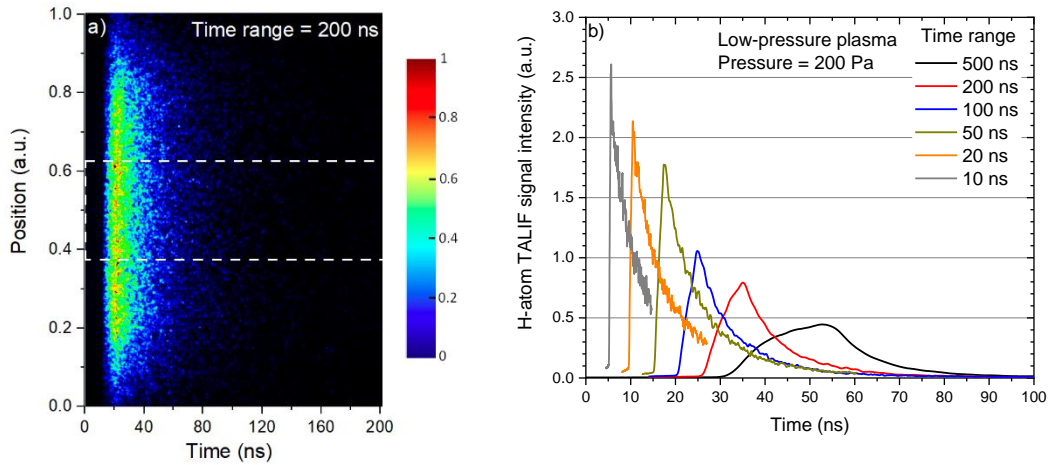


Figure 2: (a) Representative streak image showing the recorded H-atom TALIF signal in a MW low-pressure plasma (200 Pa), exemplarily shown for TR=200 ns. (b) H-atom TALIF profiles obtained from different streak measurements (spatially-averaged over the region between the dashed lines, like shown in (a)) for different TR. These curves are intentionally shifted in time for a better visualization.

Figure 2(b) illustrates that raw TALIF signals can be fully captured by the streak camera only when using $TR \geq 100$ ns. Specifically, we can identify two signal regimes which are a fast rising slope followed by a much slower exponential decay. The areas under the full signals are $(1.28 \pm 0.28) \times 10^9$, $(1.24 \pm 0.30) \times 10^9$ and $(1.23 \pm 0.39) \times 10^9$ a.u. for TR = 500, 200 and 100 ns, respectively. The small difference between these areas is mainly due to a small decrease of the laser energy during each TAT (≈ 200 s). In fact, a laser energy fluctuation of 10% over 300 laser pulses (i.e., $60 \text{ s} \approx \text{TAT}/3.3$) is systematically measured in our experiments.

As can be seen in Figure 2(b), the overall shape of the TALIF signals significantly changes with increasing TR. As a matter of fact, the rise time of the H-atom TALIF signal in Figure 2 is strongly increased by the variation of the time range of the streak camera. The rise times of the TALIF signal corresponding to each TR are given in Table 1. This distortion is similar to that observed when recording a fast signal with an oscilloscope whose bandwidth is much lower than that of the signal. On the other hand, although the short TR yield better approximations of the rising and early decay phases of the TALIF signal, they do not allow capturing the full fluorescence decay time. One may think that the determination of the actual TALIF signal can be achieved using several smaller TR windows to record the different TALIF portions along the signal's time span and to reconstruct the full signal through a "stitching" process. Unfortunately, this is not feasible due to relatively large fluctuations of the laser energy and the plasma (e.g., emission intensity for the plasma jet) when using multiple small time-ranges instead of a single one. These factors along with the difficulty to identify and set an appropriate starting/ending time for consecutive TR along the TALIF time span (due to the jitter of the delay generator), induce

undesirable variations on the fluorescence intensity making the “stitching” process” impossible. Therefore, proper attention is required towards the significant distortion of the fluorescence signal shape as a function of the TR as seen in Figure 2.

Table 1: Influence of the time range of the streak camera on the rising front duration (expressed in ns and pixels) of the H-atom TALIF. The rise time is measured as the difference between the instants where the TALIF intensity is 10% and 90% of its peak value.

Time range of the streak camera (ns)	500	200	100	50	20	10
H-atom rise time (ns)	14.27	6.60	3.39	1.73	0.81	0.46
H-atom rise time (pixels)	29	36	38	36	38	33

Consequently, a large TR will have poor temporal resolution which will artificially stretch both the rising and the exponential decay phases. However, the rising slope is more affected due to its ≈ 10 ps rise time connected to the two-photon excitation as clearly shown in Table 1. Interestingly, no difference is observed when converting the rise time in pixels (≈ 35 px in average). This is due to the constant slit width of $150 \mu\text{m}$ used for these TR. Therefore, the entrance slit width is also a crucial parameter for setting the temporal resolution of the streak camera. However, for weak fluorescence signals a trade-off between temporal resolution and SNR needs to be considered, since the intensity of the signal is directly proportional to the entrance slit width. In conclusion, there is a significant alteration of the actual fluorescence profile that must be taken into consideration when capturing ps-TALIF signals with a streak camera. The overall effects leading to the distortion of the TALIF signal can be treated by introducing an instrumental function, similar to the instrumental broadening of an optical spectrometer [22]. The raw TALIF signal collected with the streak camera can be considered as a convolution between its instrumental function and the actual TALIF signal. Since in our case TALIF signal has both ultrafast and fast components for the rising front (few ps) and the decaying part (few ns or hundreds of ps) respectively, the choice of TR as well as the slit width is critical for proper extraction of the actual fluorescence decay times. Therefore, a combination of time range and slit width has to be chosen that allows capturing the full raw TALIF signal with decent SNR.

The instrumental function of the streak camera in our case is experimentally determined using the ultrafast laser pulse as a reference signal. Specifically, we recorded with the streak camera the Gaussian temporal profile of the laser pulse and measured the variation of its FWHM as a function of the time range. For this purpose, the light from the picosecond laser is recorded with the streak camera (only a laser beam reflection is sent into the entrance slit of the streak camera to avoid damage to the camera due to excess light). The jitter of the laser trigger is only a few ps, while that associated with the streak camera and the delay generators used is estimated to be around 50 ps. This prevents in principle the use of the lowest time range provided by the fast sweep unit (i.e., 100 ps) as the laser pulse would easily exit the effective CMOS sensor area. Moreover, a dedicated mathematical process provided in the camera’s software allows for a correction of the time-variation of the appearance of the signal in the CMOS sensor due to the jitter [23]. Therefore, the laser pulse temporal profile is recorded using a TR of 200 ps (see Figure 3(a)), and not with a TR of 100 ps. After applying this process, the measured reference laser profile has a FWHM of 9.85 ps obtained by fitting the experimental raw signal with a Gaussian function. This value is in excellent agreement with the laser’s manual indicating a pulse duration of about 10 ps at 1053 nm. To investigate the effect of the time range of the streak camera on the measured FWHM of the laser pulse, the laser pulse is recorded using different TR between 200 ps and 500 ns. As mentioned before, besides the TR, the slit width of the streak camera also significantly affects the temporal resolution of the detector. Since the laser pulse is characterized by a large emission intensity (i.e., high SNR), the measurements in Figure 3 are performed with a minimum slit width of $10 \mu\text{m}$ to achieve the best possible temporal resolution and avoid any additional distortion of the temporal profile of the laser pulse. On the other hand, the choice of a slit width of $150 \mu\text{m}$ for recording TALIF signals in this work is based on the need of ensuring a good SNR while capturing fluorescence signals in both plasmas.

Figure 3(b) depicts the measured FWHM of the laser pulse as a function of the time range. The FWHM linearly increases with the increment of the time range. The recorded signal which represents the convolution between the actual laser signal and the instrumental function of the streak camera, is strongly affected by the latter for TR above 200 ps. This means that the actual laser signal can be accurately inferred from the recorded signal only for time ranges below 200 ps. The distortion of the temporal profile of the laser pulse with increasing TR is further clearly illustrated in Figure 3(c) and Figure 3(d) using representative TR values (0.5 - 200 ns). For each TR studied in Figure 3, a Gaussian function was used to approximate the temporal profile of the laser pulse. To reliably record the full TALIF signals in this work, two different TR of the slow sweep unit, i.e., 100 and 10 ns, were used for

each of the two investigated plasmas, i.e., the low-pressure plasma and the atmospheric-pressure, respectively. The corresponding instrumental functions are defined by Gaussian FWHM of (0.20 ± 0.02) ns and (3.40 ± 0.45) ns, respectively. Our choice of these two TR values (10 and 100 ns for the atmospheric- and the low-pressure plasma respectively) is justified hereafter.

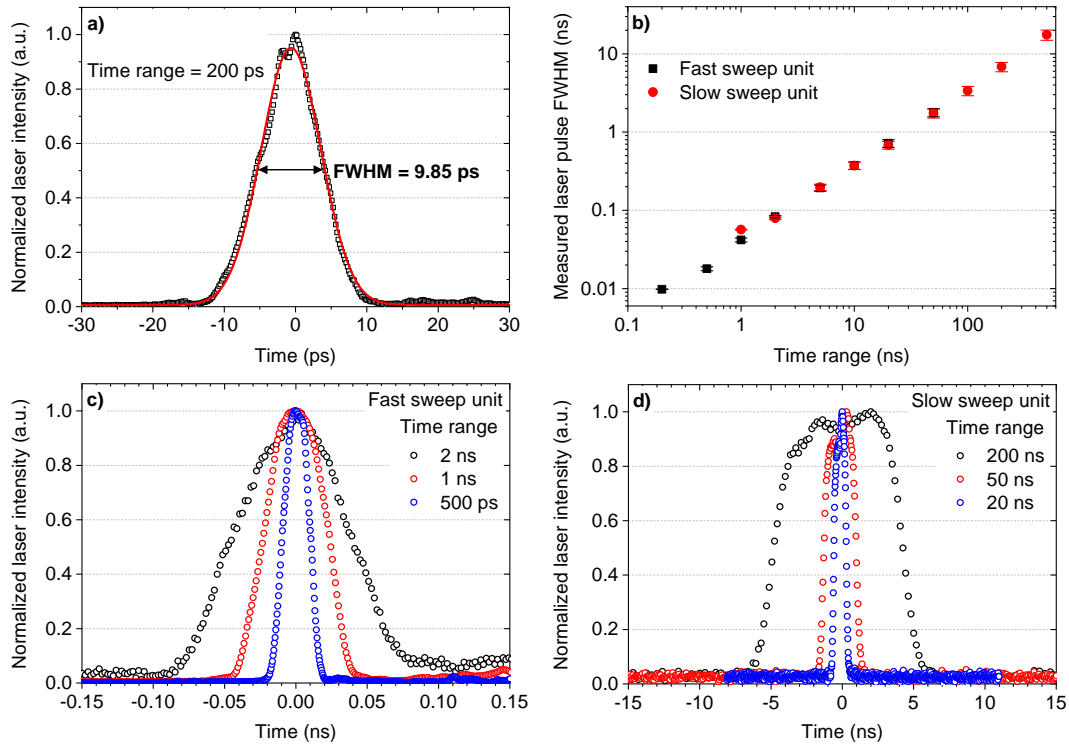


Figure 3: (a) Actual temporal profile of the laser pulse (black squares) recorded by means of the streak camera with the fast sweep unit using a time range of 200 ps and fitted with a Gaussian function (red). (b) Measured FWHM of the laser pulse versus the time range of the streak camera (both the fast and the slow sweep units were used to vary the time range between 200 ps and 500 ns). Representative measured laser pulse profiles referring to TR=0.5, 1, 2 ns (fast sweep unit) and TR=20, 50, 200 ns (slow sweep unit) are illustrated in (c) and (d) respectively. All these measurements were performed with 10 μ m slit width of the streak camera.

Furthermore, the effect of the entrance slit width of the streak camera on the temporal resolution of the streak detector is visualized in Figure 4. This figure shows the relation of the slit width and its respective width on the CMOS sensor recorded in the so-called “Focus Mode” of the streak camera (by diffuse-illumination of the slit). In this mode, no sweeping voltage is applied in the streak tube, i.e., the spatial intensity profile on the CMOS sensor is directly connected to the width of the entrance slit. The inset in Figure 4 shows the widths of the profiles (taking the total width of the profile in px, i.e., when the signal is securely above the noise level) as a function of the slit width. Since during the sweep in an actual streak measurement the spatial axis is converted to a time axis, the widths of the intensity profiles on this axis are corresponding to a temporal resolution, i.e., the px value is converted to ns. This was done exemplarily for a 100 ns time window shown as the top axis of Figure 4 (since the slit covers only a minor part of the sensor, the axis features a break to show the complete range). In addition, it is clear from the profiles shown in Figure 4 that the intensity is decreasing when reducing the slit width: the selection of a proper slit width is always a trade-off between intensity and temporal resolution.

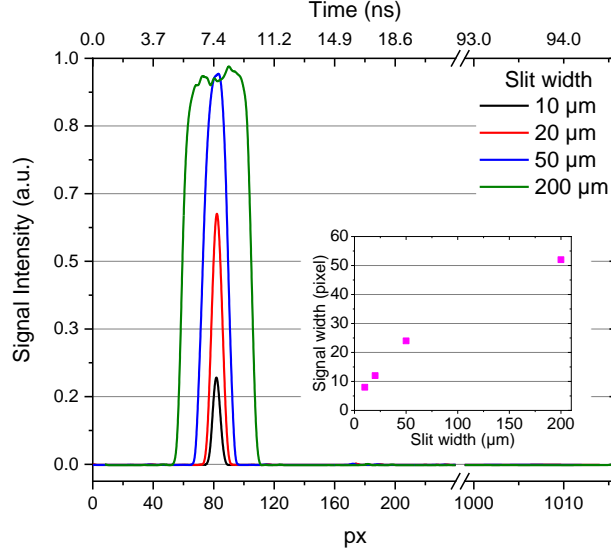


Figure 4: Visualization of the impact of varying entrance slit width on the temporal resolution using the spatial profiles of a diffuse-illuminated slit without the conversion of spatial information to time. This px-to-time conversion was exemplarily performed for a time range of 100 ns. The inset shows the total width of the profile in px, when the signal is securely above the noise level for better comparison.

Consequently, the measurements in Figure 3 are performed with a minimum slit width of 10 μm to achieve the best possible temporal resolution and avoid any additional distortion of the temporal profile of the laser pulse. On the other hand, the choice of a slit width of 150 μm (maximal meaningful value, since this is the actual height of the photocathode) for recording TALIF signals in this work is based on the need of ensuring a proper SNR while capturing fluorescence signals in both plasmas.

3.2. Interpretation of the fluorescence signals

In section 3.1 we show that the TALIF signal recorded by the streak camera consists of a fast rising slope followed by a much slower exponential decay. More precisely, the TALIF process is characterized by three distinct time regimes:

- (i) a temporal interval before the shooting of the laser pulse represented by the background noise;
- (ii) an ultrafast rising front due to the two-photon excitation process of ground-state atoms covering the laser pulse width (FWHM \approx 10 ps in Figure 3(a));
- (iii) an exponential decay starting after the previous regime and lasting over a much longer period compared to the laser pulse width.

The intensity of the raw fluorescence signal (S_F) entering the streak camera (i.e., falling on the photocathode of the streak camera) can be approximated by a simple analytical function describing the previous three regions [6]. This function, hereinafter referred to as S_{FM} , consists of the following equations: (2.1) describing the baseline of the TALIF signal (denoted A_0 ; mainly originating from the background noise and lasting up to an instant t_0 where the two-photon absorption takes place; see Figure 5), (2.2) describing a steep rising slope (A_1) which corresponds to an ultrafast increase of the TALIF between t_0 and t_1 (t_1 is the instant of the peak intensity of TALIF and $t_1 - t_0 \approx$ 10 ps (laser pulse width, FWHM)), and (2.3) defining an exponential decay from t_1 to the end of the signal, whose time constant is denoted as τ_X . These equations are formulated as follows [6]:

$$S_{FM} = \begin{cases} A_0, & t < t_0 & (2.1) \\ A_0 + A_1(t - t_0), & t_0 < t < t_1 & (2.2) \\ A_0 + A_1(t_1 - t_0) \exp\left(-\frac{t - t_1}{\tau_X}\right), & t_1 < t & (2.3) \end{cases}$$

The assumption of a linear step rising regime is acceptable in the case of ps-TALIF signals owing to the much smaller laser pulse width when compared to the time constant of the exponential decay.

However, due to the limitations of the acquisition system, the signal is modified by the Gaussian profile of the instrumental function of the streak camera as follows:

$$G = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(t-\mu)^2}{2\sigma^2}\right) \quad (3)$$

Where μ and σ are the mean and the standard deviation values of the instrumental function, respectively. As a result, the registered final raw TALIF signal with the streak camera is considered as the convolution between the actual TALIF (equations (2.1), (2.2) and (2.3)) and the instrumental function of the detector (equation (3) and Figure 3(b)) i.e., $\overline{S}_F = S_{FM} * G$. Thus, the resulting raw TALIF signal that is recorded by the streak camera becomes:

$$\begin{aligned} \overline{S}_F = A_0 + A_1 & \left\{ \frac{\sigma}{\sqrt{2\pi}} \exp\left(-\frac{(t-t_0-\mu)^2}{2\sigma^2}\right) - \exp\left(-\frac{(t-t_1-\mu)^2}{2\sigma^2}\right) \right. \\ & + \frac{(t-t_0-\mu)}{2} \left(\operatorname{erf}\left(\frac{(\mu+t_1-t)}{\sqrt{2}\sigma}\right) - \operatorname{erf}\left(\frac{(\mu+t_0-t)}{\sqrt{2}\sigma}\right) \right) \\ & \left. + \frac{(t_1-t_0)}{2} \exp\left(\frac{\sigma^2}{2\tau_x^2} - \frac{t-t_1-\mu}{\tau_x}\right) \operatorname{erfc}\left(\frac{t_1-t+\mu+\frac{\sigma^2}{\tau_x}}{\sqrt{2}\sigma}\right) \right\} \quad (4) \end{aligned}$$

In equation (4), erf is the error function and erfc is the complementary error function. It has to be noted that the convolution between an exponential decay function and a Gaussian profile is nothing but the Exponentially Modified Gaussian (EMG) function. The only difference between equation (4) and the EMG function is the contribution from the linear rise region of the fluorescence. As the TALIF signal rise time is much smaller compared to its decay time, the signal may as well be approximated by a modified exponential decay function. However, for the sake of completeness, we will use equation (4) as the form of the recorded raw TALIF signal with the streak camera. Moreover, the temporal integral of the TALIF signal does not change with the convolution between the normalized instrumental function and the raw TALIF signal. Therefore, the temporal integral of the TALIF signal can be simply expressed as:

$$\int_0^{\infty} S_{FM}(t) dt = \int_0^{\infty} \overline{S}_F(t) dt \quad (5)$$

The raw captured TALIF from the streak camera is a 2D image whose X-axis represents the spatial variation of the signal while the Y-axis represents its temporal variation (i.e., inversed axes in Figure 2(a)). The TALIF signals falling on the central region of the CMOS sensor are used to determine the spatially-averaged temporal profiles (see Figure 2(a)) and the corresponding standard deviation. Figure 5 shows representative examples of spatially-averaged temporal profiles (black open circles) along with their standard deviations (gray zones) for the low-pressure microwave plasma (at 40 and 200 Pa) and for the atmospheric-pressure plasma jet. The mean temporal signals are subjected to least-square fitting using equation (4) to obtain the different signal parameters (i.e., A_0 , A_1 , τ_x). The noise of the temporal signal averaged over the spatial dimension of the CMOS sensor, i.e., its standard deviation, has been incorporated into the least square procedure through the instrument weighting of the signal [24]. This allows for an improvement in the confidence of the fit of noisy data. In the fitting procedure, the σ value is assigned to the experimentally determined instrumental function corresponding to the TR. Since the choice of μ value of the Gaussian profile does not have a significant impact on the retrieved parameters, as the laser pulse width is much smaller than the TALIF decay time, the μ value has been fixed at t_0 coinciding with the beginning of the steep rise front of the TALIF signal. The value of μ only induces a time shift on the deconvoluted curve, which does not affect the decay time and the integral under the TALIF signal. Examples of the raw signals S_F (black circles) and their fitted curves \overline{S}_F (equation (4); red) are illustrated in Figure 5. Also shown are the constructed actual TALIF signals S_{FM} (equations (2.1), (2.2) and (2.3); blue) and the Gaussian instrumental functions G of the streak camera (equation (3); green).

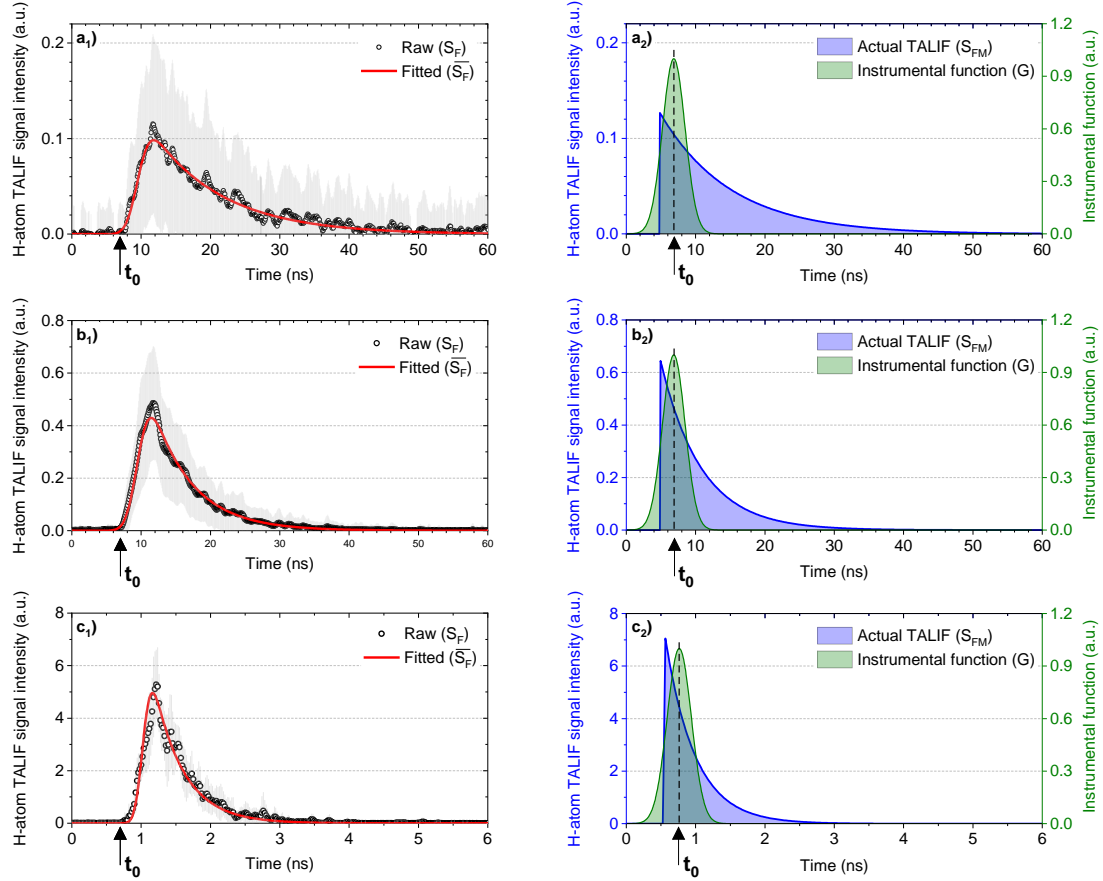


Figure 5: Raw H-atom TALIF signals S_F (black circles) recorded with the streak camera in the low-pressure microwave plasma at (a₁) 40 Pa and (b₁) 200 Pa (TR=100 ns for both), and (c₁) in the atmospheric-pressure plasma jet (TR=10 ns). These are fitted with theoretical curves \bar{S}_F (red) based on the procedure developed in this work (equation (4)). From the features of the theoretical curves, a reconstruction of the actual TALIF signal S_{FM} (blue) is performed ((a₂), (b₂), and (c₂) respectively). As the fitting method considers the weighted error of each point of the raw curve, the corresponding standard deviation is plotted as well (light gray). The Gaussian instrumental functions (G) are also shown (green) whose FWHM is 0.20 ns and 3.40 ns for the atmospheric-pressure and the low-pressure plasma respectively.

Figure 6 depicts the influence of the time range duration of the streak camera on the measured decay time of the H-atom TALIF signal in the case of a low-pressure plasma at 200 Pa (Figure 6(a)) and an atmospheric-pressure plasma jet (Figure 6(b)). These values are obtained from the fitting procedure applied to different weighted averaged signals such as those illustrated in Figure 5. It must be noted that the H-atom decay time here refers to the decay time of the laser-excited state of hydrogen (H*; n=3) generated by the absorption of two UV-photons (205.08 nm each).

It can be seen in Figure 6(a) (low-pressure plasma) that τ_H is significantly rising from 2.2 to 5.7 ns, which corresponds to a TR increase from 10 to 50 ns, respectively. Then, τ_H reaches a plateau at about 6.5 ns for TR=100 and 200 ns, while it becomes even larger for TR=500 ns. In Figure 6(b) (atmospheric-pressure plasma jet), τ_H reaches an average value of about 0.44 ns for TR ≤ 20 ns, while it almost doubles at 50 ns and becomes about 2 times larger at 100 ns. These observations clearly demonstrate the effect of the instrumental function of the streak camera as well as the noise on the extraction of H-atoms fluorescence decay times from the raw TALIF signal. For the calculation of the correct decay times from the recorded TALIF signal, its broadening by the instrumental function of the streak camera has to be taken into account. The component of τ_H due to the instrumental function decreases with TR until it becomes negligible allowing us to reach the true value of the τ_H corresponding to the decay of the laser-excited states of H-atom. This explains the plateau for both pressures. However, the extraction of τ_H is limited by the quality of the signals as well as the ability to capture the full raw signal as seen at low pressures for TR < 50 ns. Based on the above description, three regions of τ_H variations are identified, as described below.

Region I: this region is the most suitable for recording the raw H-atom TALIF in both plasmas in our conditions. It includes moderate TR values indicated by the green hatched zones in figure 5. In these zones, not only the full raw TALIF signals fit in the CMOS sensor of the streak camera, but also the extraction of the most reliable approximations of the actual TALIF signals is feasible. This is because the instrumental function of the streak camera is the smallest possible without cutting off any of the signal. However, even within this optimal region the interference with the actual TALIF is still noticeable if one compares the red and blue signals in Figure 5. The difference between the raw and actual TALIF shapes is revealed by applying the mathematical procedure proposed in this study.

Region II: lower TR values which are not sufficient for capturing the full TALIF signal (i.e., <100 ns and <10 ns referring to low- and atmospheric-pressure plasmas respectively). The temporal integral cannot be directly determined from the raw signals in these regions. However, it could still be extracted from the fitting of the raw TALIF signal using equation (4). The negative consequence of using the fitting method would be a possible error on H-atom density measurements due to inaccurate extraction of actual TALIF decay times. At low pressures, extraction of τ_H is difficult (see, e.g., Figure 6(a)) owing to the higher noise levels (SNR<50). On the other hand, an extraction of more consistent τ_H values is possible for the atmospheric-pressure plasma jet (better SNR, due to higher H-atom densities). Consequently, the temporal integrals are much more consistent than in the case of a low-pressure plasma. Nevertheless, one must avoid this region because of the possible uncertainties in the measurement of TALIF decay times and, thus, the atomic densities.

Region III: higher TR values (i.e., >200 ns and >20 ns referring to low- and atmospheric-pressure plasmas respectively). Although the full TALIF signal can largely fit in the CMOS sensor or the streak camera, the distortion of the actual TALIF by the instrumental function of the streak camera becomes significant, thus, making the extraction of the actual TALIF decay times through our fitting procedure difficult. Indeed, the actual fluorescence decay time becomes similar to or greater than the FWHM value of the instrumental function of the streak camera. For instance, at TR=500 ns (Figure 6(a)) and TR=100 ns (Figure 6(b)), τ_H/σ becomes $7.8\text{ns}/6.37\text{ns}=1.22$ and $0.98\text{ns}/0.81\text{ns}=1.21$, respectively. This region should be also avoided for recording the raw TALIF signals.

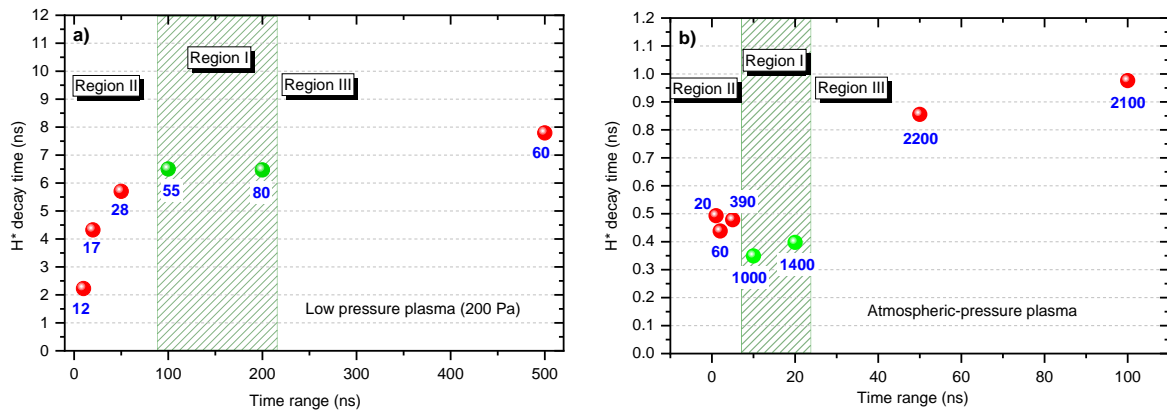


Figure 6: Effect of the time range duration of the streak camera on the measured fluorescence decay time of the laser-excited H-atom (H^*) (τ_H) in (a) a low-pressure plasma at 200 Pa and (b) an atmospheric-pressure plasma jet. The numbers appearing close to the symbols in both figures refer to the SNR value of each time range.

Based on the above analysis, it can be concluded that for a reliable extraction of fluorescence decay times when performing ps-TALIF experiments combined with a streak camera, one must choose Region I. In conditions where Region I cannot be achieved, Region II could be an alternative compromise provided the raw TALIF signal has a good SNR and a sufficiently long decay. In the present work, the following time ranges from Region I were chosen to measure H-atom decay time: TR=100 ns for the low-pressure plasma and TR=10 ns for the atmospheric-pressure plasma jet.

For investigating the sensitivity of our method, first the pressure in the low-pressure MW-driven discharge was varied between 40 and 200 Pa (5-fold increase) and the corresponding decay times of the laser-excited state of H-atom (i.e., H^* ; $n=3$) were extracted. This pressure increment already reveals a 1.6-fold decrease in the decay time of hydrogen atoms (i.e., from 10.2 to 6.5 ns according to our measurements) due to a relatively higher collisional frequency at 200 Pa compared to 40 Pa. Second, the method is further considered for TALIF experiments performed in an atmospheric-pressure plasma jet, which represents a reactive medium with a much higher collisional frequency, since its operating pressure is 500 times larger compared to 200 Pa. Thus, a much faster quenching of the laser-excited state is expected in this plasma source. Indeed, our measurements reveal that the

corresponding H-atom decay time is drastically decreased to 0.35 ns, i.e., about 19-fold decrease with respect to the value obtained at 200 Pa in the low-pressure plasma, which is in a relatively good agreement with reference [21]. Therefore, our TALIF data treatment method can be applied for extracting laser-excited H-atom decay times in different reactive plasmas operating at least in the range between 40 Pa and atmospheric pressure. Furthermore, the measurement of H-atom densities in both plasmas could be done with the deconvolution method and the good choice of TR, but this is out of the scope of this work.

4. Conclusion

In this work, we used a streak camera for capturing ps-TALIF signals of H-atoms in low- and atmospheric-pressure plasmas. It was shown that the use of the streak camera is beneficial for effectively resolving the decaying phase of the fluorescence signals in both plasmas. This was achieved by exploring the effect of the time range of the streak camera on the properties of the raw TALIF signals (shape, rise time and decay time). Using a time range that is much higher than the duration of the fluorescence signal resulted in a heavily distorted raw TALIF envelope due to the contribution of the instrumental function of the streak camera (Gaussian profile; measured by capturing the laser pulse profile with the streak camera), which changes with the time range. Thus, to record full raw TALIF signals that better approximate the actual TALIF signals we used the smallest possible time range (100 and 10 ns for low- and atmospheric pressure plasmas respectively). Although this choice allowed maintaining the instrumental function of the streak camera as small as possible, still there was a need for removing its contribution so as to obtain the actual TALIF signals. This was only possible after the application of a dedicated mathematical method involving an exponentially modified Gaussian function. The consideration of this method led to reliable measurements of decay times of hydrogen atoms in both plasmas. In fact, the choice depends on the intensity of the signal under investigation, thus, the slit width must be as low as possible to ensure a high temporal resolution while maintaining a decent signal-to-noise-ratio. This work underlines the remarkable potential of using a streak camera to capture ultrafast light signals, as exemplarily demonstrated for H-atom ps-TALIF in two different plasmas.

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