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Imaging atomizing sprays with high visibility using two-photon fluorescence laser sheet imaging

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Abstract

Two-photon excitation laser induced fluorescence (2p-LIF) is used here for imaging through an optically dense spray system. The main advantage of the approach is that a low level of unwanted fluorescence signal originating from multiple light scattering is generated. This leads to high visibility and image contrast even through scattering media, thus, providing faithful descriptions of the imaged fluid structures. While 2p-LIF imaging is a well-known point measurement approach in the field of life science microscopy [1], it has, to the best of the authors' knowledge, never been tested for observing atomizing sprays. We take advantage of this process, here, at a macroscopic scale by imaging a light sheet of ~1cm height. To generate enough 2p-LIF signal at such large scale and for single-shot detection, ultra-short laser pulses of high pulse energy are needed. This is obtained by using a laser system providing 25 fs pulses centered at 800 nm wavelength and having 2.5 mJ pulse energy. The technique is demonstrated by imaging a single spray plume from a 6 holes commercial Gasoline Direct Injection (GDI) system running at 200 bars injection pressure. The injected liquid is water mixed with Fluorescein dye. We show the important image contrast improvement of 2p-LIF light sheet imaging in comparison with the traditional shadowgraphy, laser sheet Mie scattering and back-fluorescence imaging. The proposed approach is very promising as a future tool for the detailed analysis of the dynamics of atomizing spray in the spray formation region.

Keywords: 2-photon fluorescence, multiple light scattering, high contrast imaging

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Introduction

Imaging atomizing sprays is extremely challenging due to the lack of visibility. Most often, the light intensity fraction of the non-scattered photons (the Ballistic photons) ranges from 13.5% down to 0.01%. In simple terms of visibility, this level of light transmission corresponds to the transition between "seeing a blurred object" to "not seeing it at all". In such situation a very large amount of the detected light intensity originate from photons that have been scattered multiple times; directly hiding spray features which are responsible for the spray formation.

To overcome problems related to multiple scattering, the experimental development of advanced imaging techniques and the means employed for the characterization of optically dense sprays has largely increased during the past decade. Three main imaging approaches in which multiple scattering can be mitigated currently exist: Ballistic Imaging (BI) [2], X-ray imaging [3] and Structured Laser Illumination Planar Imaging (SLIPI) [4]. Despite their merits, those techniques have various limitations: BI and X-ray techniques are line-of-sight techniques and are usually employed in the near field spray region, close to the injector tip, where large liquid structures are visible. On the opposite, thanks to its light sheet configuration, SLIPI provides spatially resolved images from large spray volumes, at locations where the spray is already formed corresponding to some distance from the nozzle tip [5]. However, when applied in the near field region, SLIPI can be limited by the presence of large irregular liquid bodies, which eliminate the incident modulated line structures due refraction from those liquid/air interfaces.

Microscopic imaging is now a popular approach for spray diagnostics, thanks to the recent development of high quality long range microscope objectives. It has been successfully applied to the study of Diesel sprays as shown in [6-7]. However, those high resolution spray images were recorded on a shadowgraphy configuration without optical sectioning advantages. Recently, it has been shown that Light Sheet Fluorescence Microscopy (LSFM) [8] could provide faithful spray information. Nevertheless, the approach remains affected by effects from multiple light scattering. Here, we propose the possibility of using a two-photon excitation scheme, instead of single-photon excitation.

The advantage of 2p-LIF detection is that it provides much higher visibility through the probed spray. The main reason for this is that photons which undergo multiple scattering processes do not carry enough energy to

induce a 2p-LIF signal. On the contrary, at the distance where the illuminating light sheet is focused the probability for the 2p-LIF process to occur is much higher; providing a signal that is only generated at the object plane of the camera objective. The efficiency of the approach is tested, here, on a 6 holes commercial GDI spray system at 200 bars injection pressure and for 50, 100 and 200 µs after the visible start of the injection. First, an image comparison is performed for various imaging approaches, including:

1) Shadowgraphy
2) Back-fluorescence imaging
3) Laser sheet Mie scattering
4) Light sheet two-photon fluorescence imaging

Then, a description of the benefits obtained when using two-photon fluorescence for analyzing in detail the near field region of the studied spray systems is given.

Two-photon excitation fluorescence

Two-photon absorption or excitation is a nonlinear process that was theoretically derived by Maria Göppert-Mayer in 1931 and later experimentally proven after the invention of the laser [9]. Electrons within an atom can be excited from the ground state to an excited state by incoming photons, on the condition that the energy of the photon matches the difference in energy between the two states. If the electron is excited by two photons instead of one, where their combined energy satisfies the energy requirement, the event is called two-photon excitation, as shown in Figure 1(b). Fluorescence signals collected from two-phase flow systems are commonly generated from single-photon excitation. However, under certain conditions, they can also be generated from two-photon excitation. In this case the energy density of the incident beam is an important parameter to increase the probability that two photons get absorbed simultaneously. Therefore, ultra-short laser pulses (e.g. ~80 femtosecond) are usually used to induce this process. The main advantage of this scheme, is that the Mie multiple light scattering from the incident radiation which induces single-photon fluorescence does not have enough energy to induce two-photon fluorescence. As a result, a large portion of undesired signal, as illustrated in Figure 1 (a) and (b), is not generated with two-photon excitation, leading to visibility enhancement. A second advantage is the possibility to sharply focus the incident light sheet to locally induce a signal and counter balance the effects of light extinction along the incident path of the laser beam. A third advantage is that any single-photon fluorescence generated at higher wavelengths than the excitation wavelength (e.g. at 800 nm) can easily be filtered out and will not interfere with the desired signal.

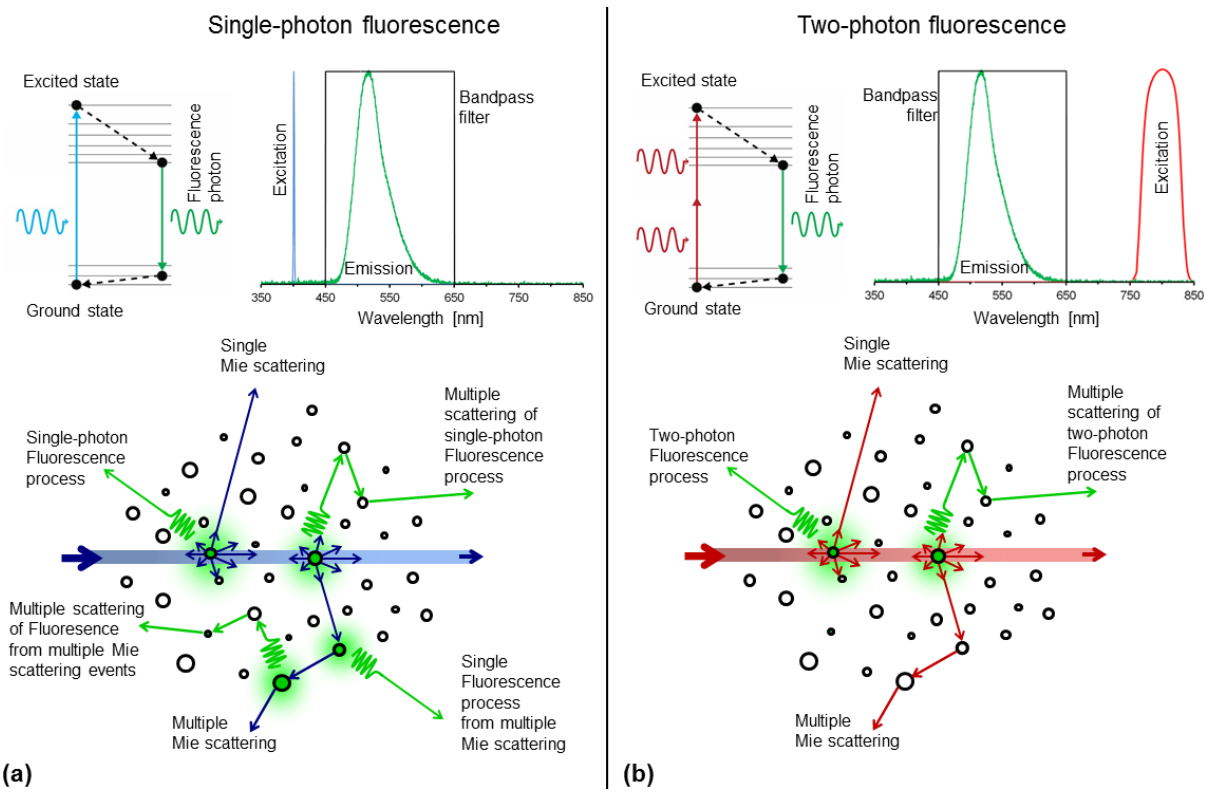

Figure 1 Comparison between single-photon (a) and two-photon (b) excitation processes. The fluorescence from two-photon excitation is only generated near the focal point of the light sheet, reducing a significant part of unwanted fluorescence outside of the light sheet.

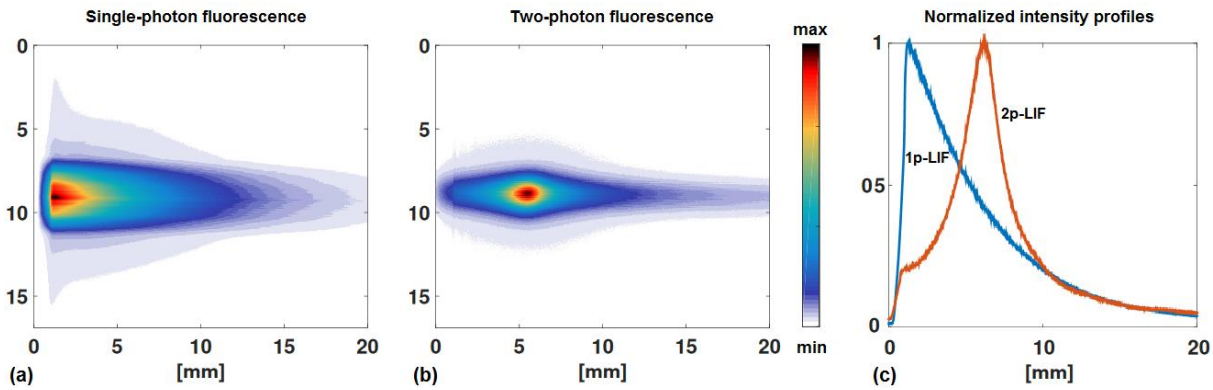

Figure 2 Comparison between single-photon (a) and two-photon (b) excitation processes together with their normalized intensity profiles in (c). Here, a 20 mm cuvette containing a homogeneous solution of water mixed with Fluorescein is illuminated with a laser beam focused with a cylindrical lens of f = 50 mm focal distance. The fluorescence signal from two-photon excitation varies significantly from the signal generated by single-photon excitation.

When illuminating a homogenous liquid volume containing fluorescing molecules, the signal generated by single-photon fluorescence is exponentially reduced with the path length x, according to the Beer-Lambert law:

$$I_{(x)} = I_0 \cdot e^{-N \cdot \sigma_a \cdot z} \qquad \text{Eq.1}$$

where I_0 is the incident light intensity, N is the number density of the dye molecules and σ_a is the single-photon absorption cross-section. This light reduction with distance can be seen in Fig.2 (a) and from the blue curve given if Fig.2 (c). However, for two-photon absorption, the generated fluorescence differs fundamentally from single-photon absorption and is described as:

$$I_{(x)} = \frac{I_0}{1 + N \cdot \delta \cdot z \cdot I_0} \qquad \text{Eq.2}$$

where δ is the molecular two-photon cross-section quoted in the units of Goeppert-Mayer (GM). By definition, 1 GM = 10−50 cm4 s photon−1, corresponding to the product of the cross-sections, in cm2, from each photon. Note that for 2p-LIF, the focal distance of the focusing lens plays an important role as shown in Fig.3. To obtain a signal homogeneously generated along the distance x a focal distance corresponding to f = 150 mm is used for imaging the GDI spray.

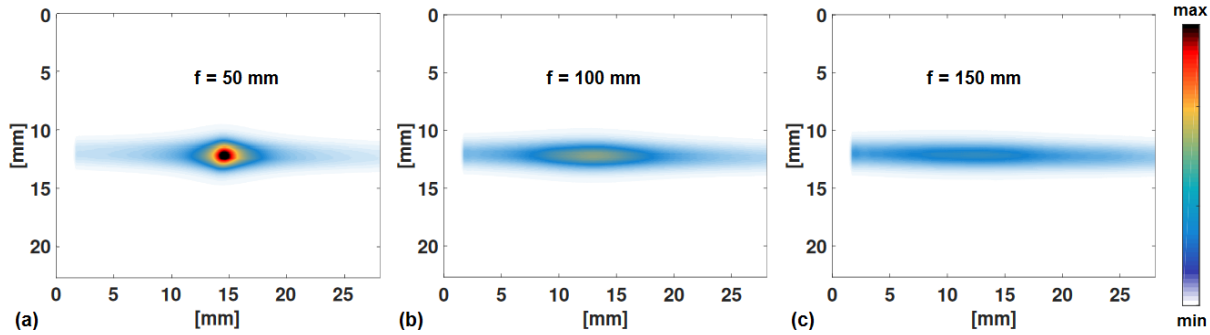

Figure 3 Effect of the focal distance of the focusing cylindrical lens for the generation of two-photon fluorescence from a homogeneous water solution mixed with Fluorescein. Here the focal distance of the focusing lens equals 50 mm, 100 mm and 150 mm, respectively. A more local and intense 2p-LIF signal is generated with shorter focal distance.

Experimental setup

The laser system is a titanium-sapphire chirped pulse amplification femtosecond system, delivering pulses at 1 kHz repetition rate, 800 nm central wavelength with a pulse energy up to 5 mJ. It comprises four amplification stages: a low-energy multi-pass preamplifier, a regenerative amplifier, and two more multi-pass amplifiers. The last amplifier is cryogenically cooled, with an output energy of up to 10~mJ. The laser system has two acousto-optic programmable dispersive filters (AOPDF) [10]. The first one (Dazzler, Fastlite) acts as a pulse shaper [11] between the oscillator and the first amplifier, and the second one is used as a programmable gain

filter [12] inside the regenerative amplifier (Mazzler, Fastlite) to reduce gain-narrowing and keep a large bandwidth. The spectral phase of the pulses is measured at the end of the amplification chain with a commercial system based on self-referenced spectral interferometry (Wizzler, Fastlite) [13] and iteratively corrected with the phase shaper (Dazzler). The pulses used for the experiment had 2.5 mJ of energy and 25fs duration, measured on target using the d-scan technique [14].

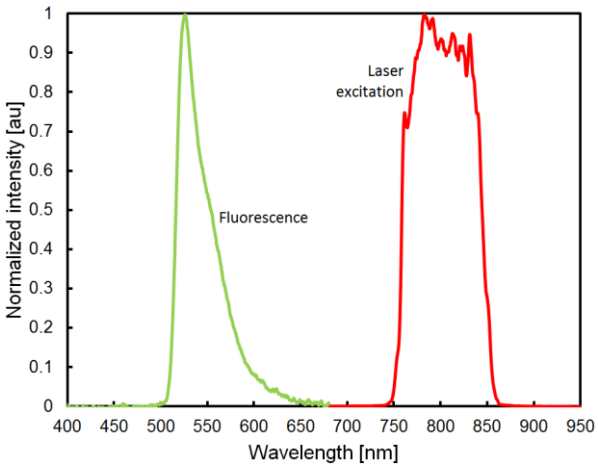

Figure 4 Spectra of the incident laser pulse, centred at 800 nm and of the resulting fluorescence signal from Fluorescein. A bandpass filter of centered at 510 nm with 90 nm bandwidth is used to only collect the fluorescence signal. The blocking of the filter corresponds to an optical density larger than 6.

The spray system is produced from a commercially available 6-holes GDI injector. The injected liquid is a solution of water with Fluorescein dye, which is prepared by mixing 40 mL solution of dye concentrated at 7% by weight within 2400mL water; pH~7. Fluorescein presents several advantages:

1) The absorption spectrum for two-photon excitation [15] matches well with the femtosecond pulse excitation used here.

2) The quantum yield of fluorescein emission in water is very high, usually >0.9 depending on the pH.

3) It is a non-toxic organic dye, which is highly soluble in water.

The liquid is injected at 200 bars pressure of injection in ambient temperature and pressure conditions. Two alternative hydraulic intensifiers are used to keep the pressure of the working fluid stable during injection. Only one spray plume is illuminated with the light sheet. The imaging system consists of a telecentric lens objective (Techspec Gold TL from Edmund Optics of x1 primary magnification with more than 50% image contrast at 40 lp/mm) mounted on a CCD camera (Luca R from Andor Technologies). In order to evaluate the performance of 2p-LIF light sheet imaging, the approach is compared with shadowgraphy and laser sheet Mie scattering. For each imaging case the exact same detection system has been used and the spray was running under identical conditions.

Results and discussions

One important aspect of the work presented here is to determine the benefits of 2p-LIF by comparing it with other optical configurations and detection schemes.

1) The shadowgraphy detection is obtained by positioning and exciting a fluorescing flat screen at the back of the spray. This configuration, allows generating a non-coherent diffuse light source.

2) The back-fluorescing signal is obtained by inserting Fluorescein in the injected water, fixing a fluorescence bandpass filter in front of the camera objective and illuminating the spray by inserting two mirrors additional mirrors.

3) The Mie-scattering signal is obtained by simply removing the fluorescence band-pass filter in front of the camera and injecting water without any fluorescing dye. Note that we define, here, “Mie-scattering” by the light being elastically scattered by any liquid elements present in the spray and not by the spherical droplets only. The light sheet is created by inserting a 150 mm cylindrical lens.

4) The two-photon fluorescence light sheet imaging setup is obtained by adding the Fluorescein dye in the water, inserting the fluorescence bandpass filter in front of the camera objective and using a 150 mm cylindrical lens to create the light sheet.

The conditions of detection were identical and the spray is generated at 200 bars pressure of injection. The images were recorded at 100 µs and 200 µs after the visible start of injection as shown in Fig.6 and Fig.7 respectively. It is seen from the zoomed areas that the liquid structures can be observed with much higher contrast with 2p-LIF than for the two other approaches.

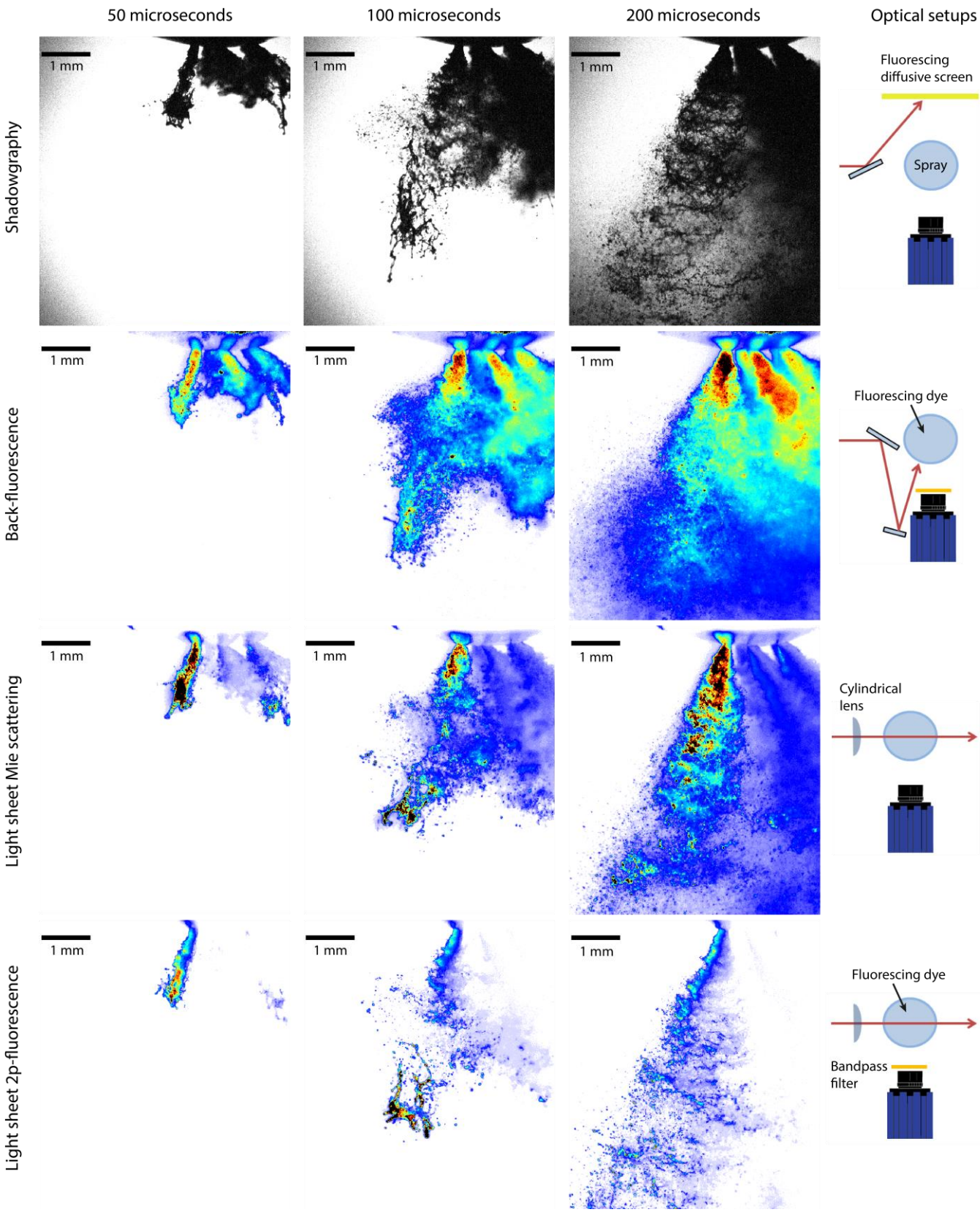

Figure 5 Image comparison between shadowgraphy, back-fluorescence, light sheet Mie scattering and light sheet 2p-LIF imaging for a GDI spray at 50 µs, 100 µs and 200 µs after the visible start of injection. Note that each image corresponds to independent injection events. In contrast with all other techniques, the presence of the other plumes is not visible when using the light sheet 2p-fluorescence approach, where individual liquid structures are visible with high contrast.

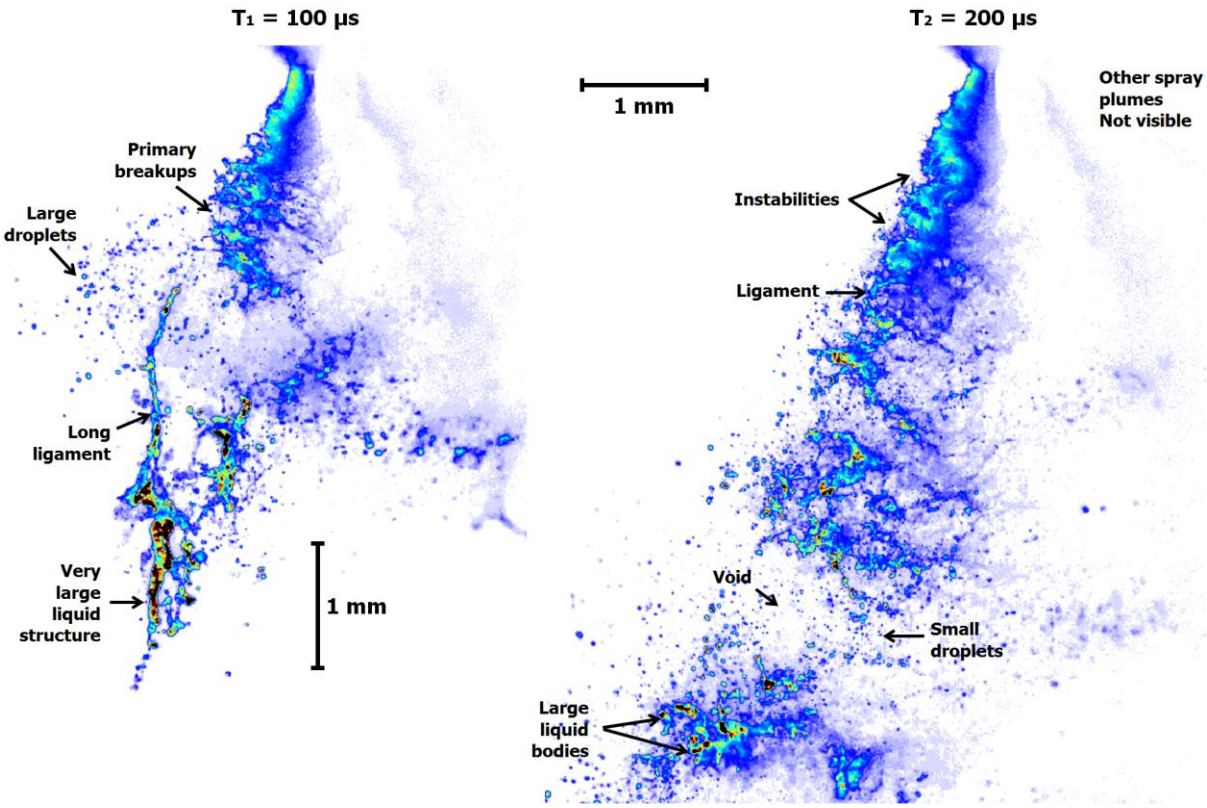

Figure 6 Image results of 2p-LIF light sheet imaging of a GDI spray at 100 µs and 200 µs after the visible start of injection.

In Figure 6, a detailed description of the spray systems for the two different times after the visible start of injection is provided. One important observation from those results, is that the other 5 spray plumes are not visible from the 2p-LIF images. This indicates that the effects from multiple light scattering, located outside from the imaged light sheet do not generate any fluorescence signal. A future work will consist in comparing those 2p-LIF spray images with 1p-LIF images.

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