

Bright-field and dark-field modes in microscopic high-speed droplet imaging

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ABSTRACT

Spray-flame synthesis is a versatile process for producing metal-oxide nanoparticles from precursor-laden liquid fuels. After atomization and ignition, the burning droplets undergo thermally induced breakup, referred to as droplet disruption, which includes puffing and micro-explosions. These events influence the release of precursor material and the formation environment of the particles. However, their experimental investigation is challenging because the droplets are small, move at velocities on the order of 100 m/s, and disrupt on microsecond time scales. Previous microscopic bright-field shadowgraphy enabled direct visualization of droplet disruption, but its temporal resolution is limited by the camera frame rate and by the loss of contrast when multiple illumination pulses are stacked within one camera frame. Furthermore, weakly deflecting small fragments are difficult to detect in bright-field images.

In this work, a combined bright-field and dark-field imaging system is developed to investigate droplet disruption in a standardized SpraySyn burner. The dark-field channel uses pulsed blue LED illumination, while the bright-field channel uses synchronized red LED illumination for reference shadowgraphy. By exploiting the dark background in dark-field imaging, 15 illumination pulses with a spacing of $2\ \mu\text{s}$ were stacked within a single camera frame, corresponding to an effective temporal resolution of 500 kHz. In comparison, three synchronized bright-field pulses were recorded within the same frame. The system was operated at 40 mm height above the burner on the burner center axis. The comparison of bright-field and dark-field recordings show that bright-field imaging provides a more intuitive representation of the overall droplet morphology, whereas dark-field imaging offers substantially improved sensitivity to weakly deflecting structures like small fragments. During a measurement campaign comprising 500 image pairs, bright-field imaging identified 56 droplet disruption events in 47 frames, while dark-field imaging identified 131 events in 88 frames. The maximum observed fragment count increased from 8 in bright-field imaging to 14 in dark-field imaging, and the average maximum visible fragment count increased from 3.4 to 5.0.

These results demonstrate that dark-field burst illumination is a promising diagnostic approach for high-temporal-resolution imaging of droplet disruption in spray-flame synthesis. The method enables improved detection of small fragments and suggests that droplet disruption may occur more frequently than inferred from bright-field observations alone.

1. Introduction

Metal-oxide nanoparticles have a wide range of industrial applications, including but not limited to magnetic materials, catalysis, and sensors (Teoh et al., 2010). Spray-flame synthesis (SFS) allows the production of these particles from low-cost precursors dissolved directly in the fuel and therefore does not require pre-vaporization, unlike conventional vapor flame reactors. (Mädler et al., 2002). The fuel-precursor mixture is atomized into droplets in the tens-of-micrometers size range, moving at approximately 100 m/s, and subsequently ignited. The droplets then undergo further breakup through thermally induced disintegration, referred to as droplet disruption. Droplet disruption can be further classified into 'puffing', a local ejection of gas and secondary droplets from a primary droplet that remains mostly intact, and 'micro-explosion', a violent fragmentation of the primary droplet. Depending on the composition and ratio of the precursor-fuel mixture, as well as the size of the individual droplet, droplet disruption occurs on the order of tens of microseconds after ignition and typically lasts less than 10 μ s itself. (Jüngst et al., 2023; Südholt et al., 2024).

Thus, the challenge here is achieving simultaneously high spatial and temporal resolution in a field of view (FOV) that is large enough to follow the fast-moving droplets during their disruption. Previous work (Stodt et al., 2020; Stodt et al., 2021; Stodt et al., 2023), including our own (Jüngst et al., 2022; Jüngst et al., 2023; Jüngst et al., 2024; Jüngst et al., 2024), used microscopic shadowgraphy. In the sparse sprays considered here, this bright-field (BF) technique yields images that are straight-forward to interpret – dark droplets on a bright background. To increase the temporal resolution, each frame recorded by the high-speed camera was illuminated multiple times by pulsing the LED light source at an integer multiple of the camera frame rate. The trade-off of this illumination scheme is a reduction in contrast, as the bright background accumulates beneath each droplet shadow. Figure 1 shows that contrast remains visually acceptable to 7 illuminations. However, automated analysis in previous work remained viable only up to three illuminations per frame (Jüngst et al., 2022; Jüngst et al., 2023; Jüngst et al., 2024).

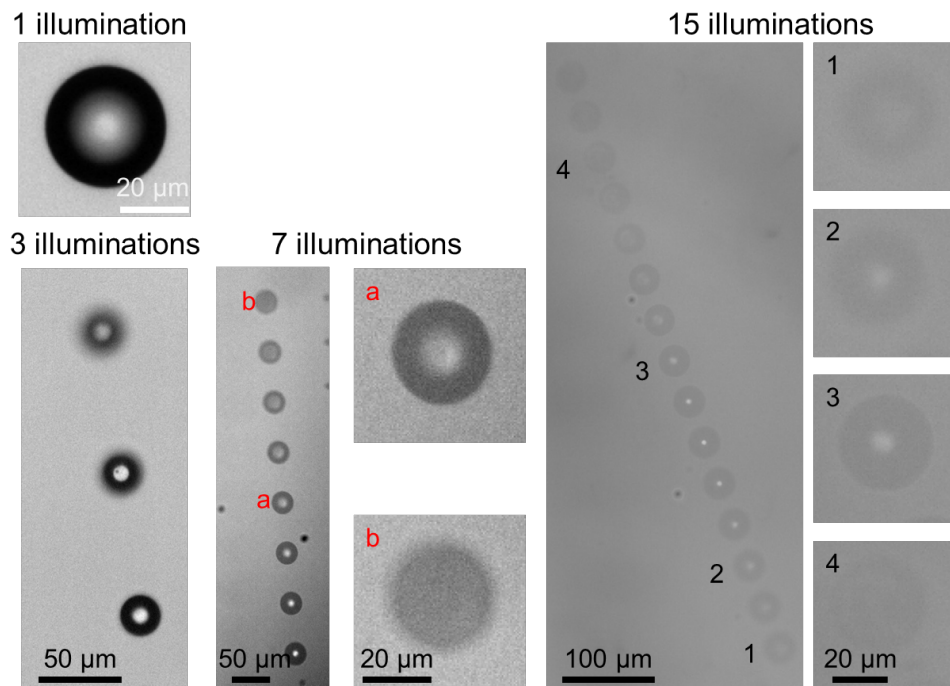


Figure 1: Contrast loss in bright-field imaging with increasing number of illuminations per frame.

In the current work, we explore options for improving multi-pulse illumination stacking. This would be useful for increasing temporal resolution or, if enough pulses can be stacked within a single frame, following the track of a droplet through the FOV in a single frame. The latter would enable replacing the high-speed camera with a much cheaper and electro-optically better “low-speed” standard CMOS camera.

The technique used here is dark-field (DF) imaging. In this imaging mode, only light deflected into the entrance pupil of the objective is detected, while undeflected transmitted light misses the objective. As a result, the background appears dark, allowing a larger number of illuminations to be stacked within a single frame, without reducing the contrast. We previously used this imaging mode, with a different optical setup, to detect aluminum oxide particles surrounding burning aluminum droplets (Jüngst et al., 2025).

2. Methods

A schematic of the experimental setup, including the position of the detection region on the burner center axis, is shown in Figure 2. The setup is based on the standardized SpraySyn burner (Schneider et al., 2019). The precursor solution is injected through a central nozzle and atomized by a surrounding high-velocity oxygen coflow. The spray flame is ignited and stabilized by a lean, premixed CH₄/O₂ flat-flame pilot. Both the pilot flame and the outer concentric air coflow, used

as sheath gas, are supplied through concentric sections of the same sintered bronze matrix to stabilize the flame environment. The characteristic spray conditions of the SpraySyn burner, including typical droplet sizes and velocities as well as Weber numbers below the critical value for aerodynamic breakup in the investigated flame regions, are summarized by (Jüngst et al., 2023). For nanoparticle synthesis, a precursor solution of 0.2 mol/L iron(III) nitrate nonahydrate (INN) was used. The precursor was dissolved in a solvent mixture consisting of 35 vol.-% ethanol (EtOH) and 65 vol.-% 2-ethylhexanoic acid (EHA). All droplet images in the results section were acquired at 40 mm height above burner on the center axis.

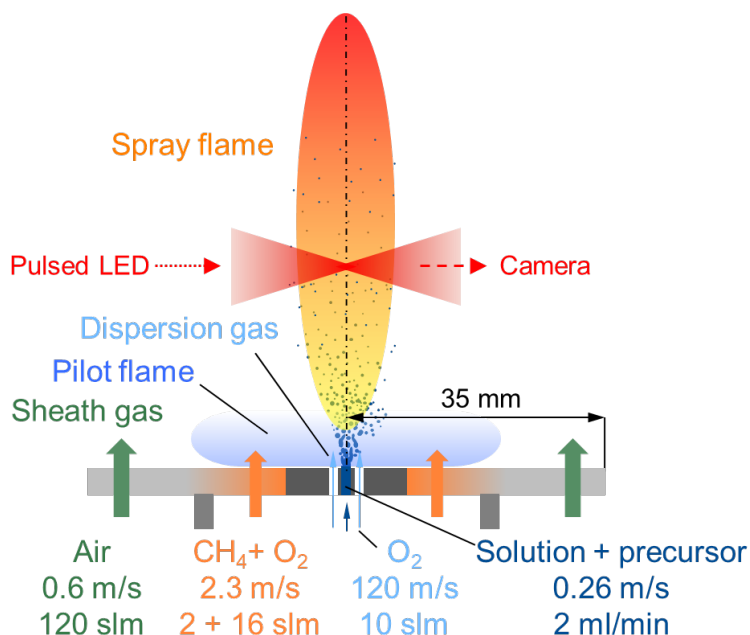


Figure 2: Sketch of standardized SpraySyn burner adapted from (Jüngst et al., 2022).

Figure 3 shows a schematic of the combined BF (red) and DF (blue) imaging setup. The BF illumination was provided by a red LED (Nichia NCSR219B-V1, peak emission wavelength 630 nm, full width at half maximum 16 nm), while the DF illumination was provided by a blue LED (Cree XLamp XP-E2 royal blue, peak emission wavelength 450 nm, full width at half maximum 16 nm). Both LEDs were pulsed for 80 ns using diode drivers (Picolas LDP-V 50-100 V3.3).

Both illumination paths were arranged in a Köhler-type configuration. The light from each LED was collimated by an aspheric collector lens with a focal length of 16 mm. An auxiliary lens with a focal length of 200 mm imaged the LED into the illumination aperture plane. An aspheric condenser lens with a focal length of 40 mm then provided homogeneous illumination of the FOV.

To achieve dark-field illumination, the center of the blue illumination beam was blocked. This was realized by stacking a 32 mm diameter bandpass filter (BP, Semrock FF01-635/18-32) and an orange glass long-pass filter (Schott OG 550). The filter stack was required because backscattering of blue light from the spray onto the bandpass filter and back into the objective lens reduced the contrast in the DF images.

The light from both illumination paths was collected by a microscope objective (Mitutoyo M Plan Apo 10x) and imaged onto the cameras (LaVision Imager sCMOS CLHS) using an achromatic tube lens with a focal length of 125 mm. This resulted in a magnification of 6.5 and a projected pixel size of $1\text{ }\mu\text{m}/\text{pixel}$.

To suppress flame luminosity from the spray flame, a bandpass filter (Semrock 447/60 BrightLine) was used in the DF channel, while a second orange glass long-pass filter was used in the BF channel. The BF and DF signals were separated using two dichroic beam splitters (BS, Thorlabs DMSP490) with a cutoff wavelength of 490 nm.

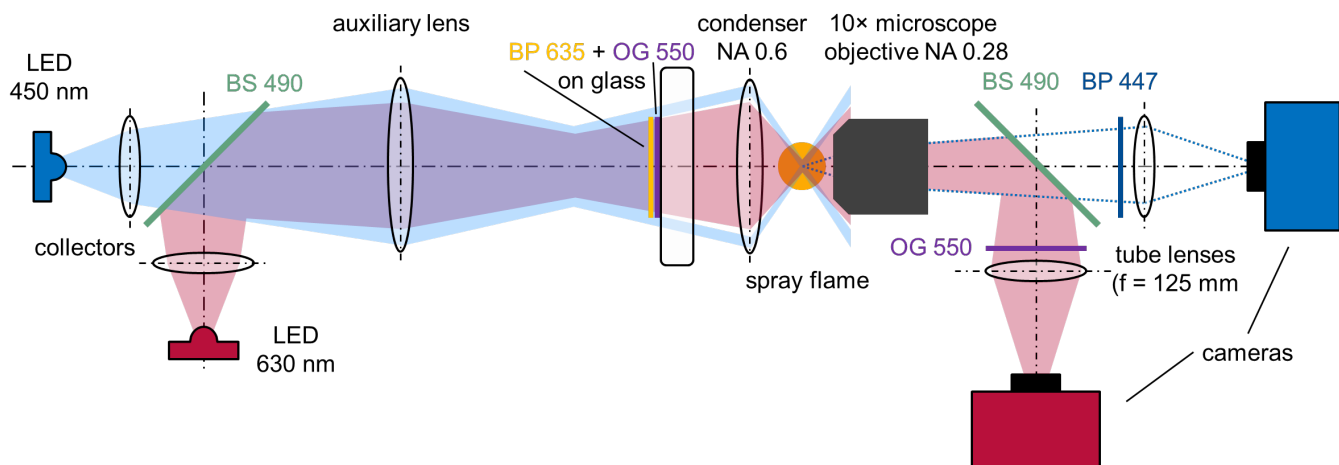


Figure 3: Sketch of optical layout viewed from the top. Dark-field in blue, bright-field in red.

The cameras had a full-frame resolution of 2560×2160 pixels but were cropped slightly to 2240×2160 pixels, resulting in a FOV of approximately 2.2×2.2 mm. The cameras were operated at 10 Hz with an exposure time of $45\text{ }\mu\text{s}$ and temporally and spatially calibrated. A spatial offset of approximately $100\text{ }\mu\text{m}$ in both the x- and y-directions was corrected during post-processing.

To increase the temporal resolution of the detection system, the blue DF LED was pulsed 15 times within a single camera frame with a pulse spacing of $2\text{ }\mu\text{s}$, resulting in an effective frame rate of 500 000 fps. To aid interpretation of the DF images, the red BF LED was pulsed three times with a pulse spacing of $10\text{ }\mu\text{s}$ and synchronized with the 1st, 6th, and 11th DF pulses. Figure 4 summarizes the illumination scheme.

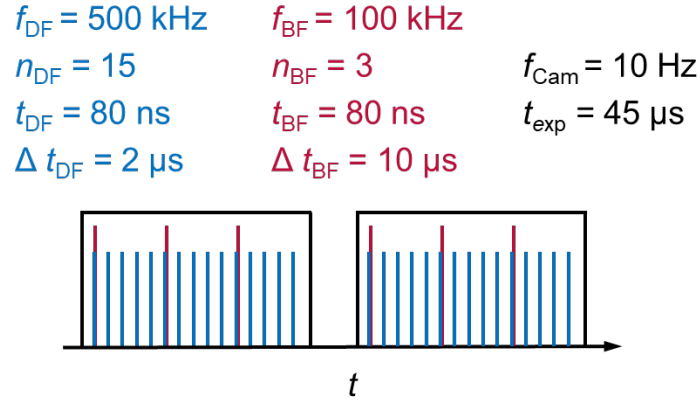


Figure 4: Timing diagram of the illumination and camera acquisition scheme. Dark-field illumination is shown in blue, bright-field illumination in red, and the camera exposure in black. Fifteen dark-field pulses and three bright-field pulses are stacked within one camera frame, with the 1st, 6th, and 11th dark-field pulses synchronized to the bright-field pulses.

Each BF image (I) was postprocessed by subtracting a constant background value (BG) and normalizing the result by the background-corrected ensemble average of all BF images in the image set, ($\bar{I}$). The resulting post-processed image (I^*) is therefore given by

$$I^* = \frac{I - BG}{\bar{I} - BG} \quad (1)$$

A direct comparison between the raw and post-processed BF images is shown in Figure 6.

3. Results

Figure 5 shows cropped, uncorrected BF and DF images of a glass SL4 Image Analysis Test Target from JD Photo Data. The target contains 14 opaque circles with diameters of 2, 4, 6, 8, 10, 12, 14, 16, 18, 20, 25, 30, 35, and 40 μm . Figures 5a and 5b show the 12, 14, 16, and 18 μm circles. The edges of the target structures appear as bright rings in DF illumination, whereas they appear as gray gradients in BF illumination. Small dust contaminations on the glass substrate, with sizes of approximately 1 μm or smaller, are clearly visible in the DF image.

Figure 5c and 5d compare the 2, 4, 6, and 8 μm circles. In the DF image, analysis of the dot morphology of these smaller objects becomes difficult because inhomogeneous DF illumination by slight misalignment strongly affects how light is deflected into the detection optics. Therefore, if accurate object size determination is required, an in-focus BF image, such as that shown in Figure 5d, is necessary.

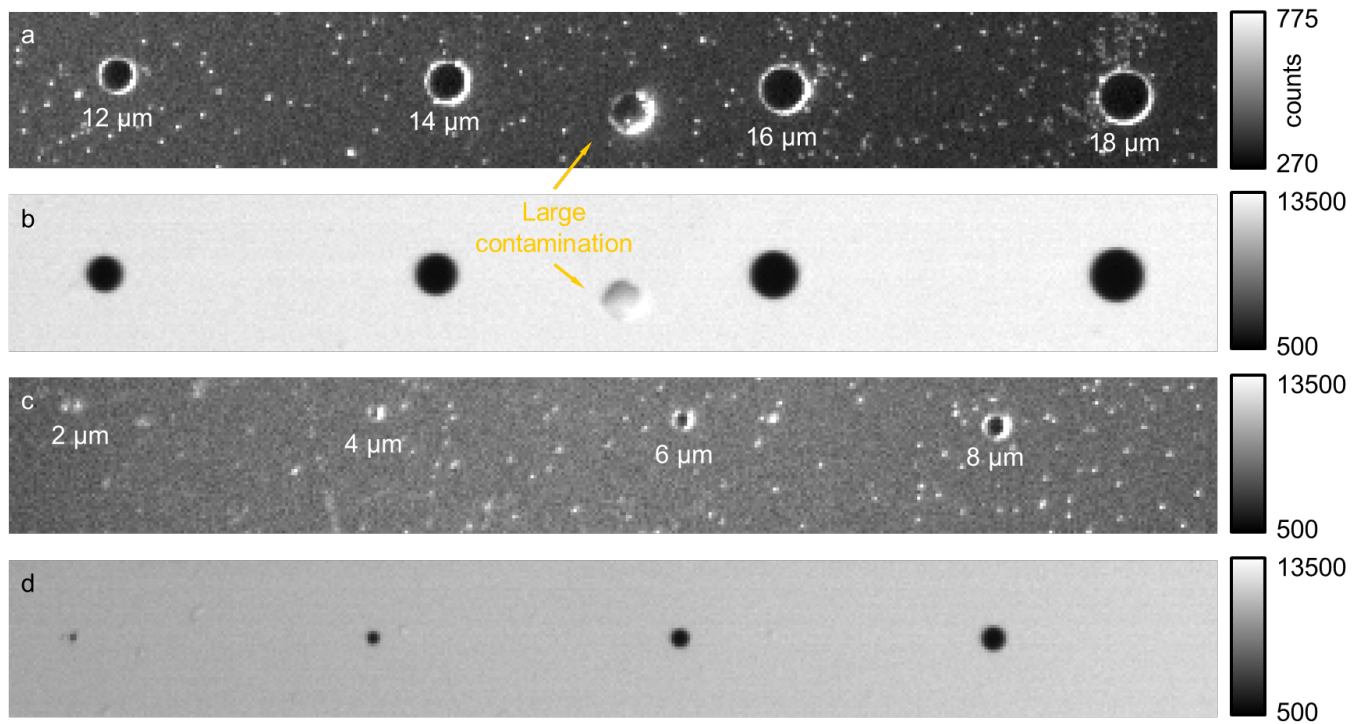


Figure 5: Opaque dot targets with nominal diameters indicated in the image, imaged using (a, c) dark-field and (b, d) bright-field illumination. The dark-field images reveal dust contamination on the glass substrate as individual bright pixels, likely originating from contamination smaller than a single image pixel. The smaller dot targets in (c) (2 and 4 μm) appear slightly larger than in (d) and exhibit increased signal intensity on their left and right sides. This effect is attributed to slight misalignment of the dark-field illumination, resulting in non-uniform illumination across the field of view.

Figure 6 shows a full-frame comparison of an uncorrected and corrected BF image in Figure 6a and 6b, respectively, together with the corresponding DF image recorded by the second camera displayed at two different dynamic ranges in Figure 6c and 6d. The corrected BF image shows substantially improved homogeneity and increased droplet contrast.

The displayed dynamic range of the DF image must be chosen depending on the features of interest. At a higher displayed dynamic range, low-intensity features disappear, while brighter features reveal additional detail. In Figure 6c, large out-of-focus droplets become visible and contaminate the apparent background. However, this lower displayed intensity range is necessary to also visualize smaller objects, since they deflect less light than larger droplets. This is particularly relevant for the detection and visualization of droplet fragments, which are much smaller than the primary droplets.

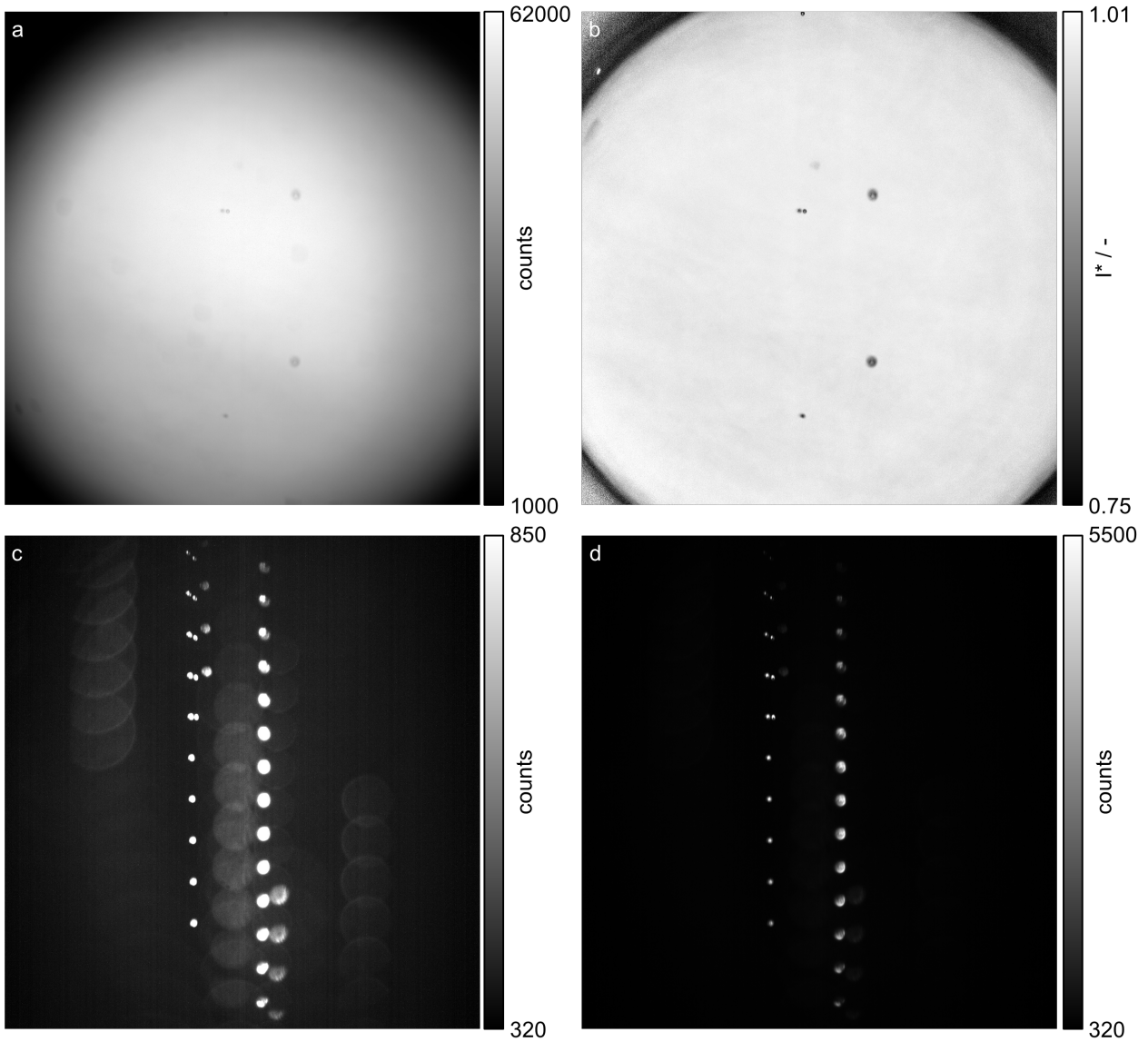


Figure 6: Raw bright-field image (a) together with the corresponding background- and flat-field-corrected bright-field image (b). Raw dark-field images shown with lower (c) and higher (d) displayed dynamic range. At higher displayed dynamic range, low-intensity features disappear, while brighter features reveal additional detail.

Figure 7 shows consecutive illuminations of a regular, non-disrupting droplet in DF and BF imaging. The BF image strip is shorter because the camera frame ends at that point at the bottom. The time axis starts at $6 \mu\text{s}$. Based on the timing diagram in Figure 4 and the position of the BF shadowgraphs, it can be inferred that the droplet was not yet within the FOV during the first three DF illumination times and the first BF illumination time.

The contrast remains good in DF imaging for all illumination times. However, the images at $6, 8, 10, 24, 26,$ and $28 \mu\text{s}$ show intensity inhomogeneities along the vertical axis. This is attributed to

the DF illumination geometry. Since the detection region is illuminated by an annular beam that overlaps with itself, the illumination intensity is not fully homogeneous and depends on the position of the droplet within the DF illumination and the FOV. As the droplet passes close to the center of the detection region, the inhomogeneity becomes visible mainly at the top and bottom of the droplet image. The cropped and enlarged BF and DF images at 10 and 20 μs show that this inhomogeneity is not present in the BF images.

In both imaging modes, slight motion blur is visible. Assuming a droplet velocity of 100 m/s and using an illumination time of 80 ns, a motion blur of 8 μm , corresponding to 8 pixels, is expected in all recordings. However, the outer morphology of the droplet is easier to interpret in BF imaging, while the droplet appears slightly larger in DF imaging.

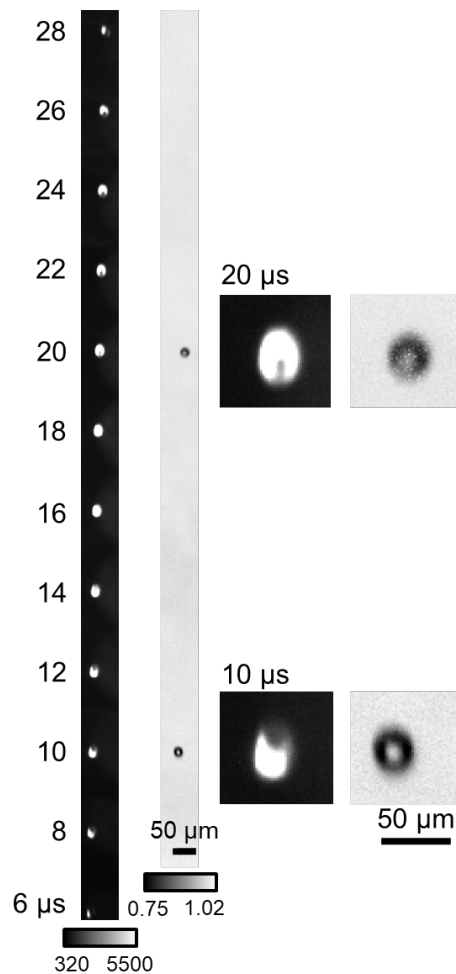


Figure 7: Dark-field and bright-field images of a non-disrupting droplet illuminated multiple times within the same camera frame using two synchronized cameras. The illuminations at 10 μs and 20 μs show corresponding dark-field and bright-field recordings.

Figure 8 shows a droplet disruption recorded over 12 DF and two BF illuminations. At $10\ \mu\text{s}$, the droplet morphology appears different in the two imaging modes. While the overall shape of the main droplet mass is easier to interpret in the BF, the DF reveals intensity modulations at the bottom right and top left of the droplet, indicating possible material ejection.

At $12\ \mu\text{s}$, the droplet undergoes further fragmentation. At $16\ \mu\text{s}$, 13 individual fragments can be identified in the DF, as indicated by the yellow numbering. These fragments exhibit varying signal intensities. Comparing the DF and BF at $20\ \mu\text{s}$, seven fragments can be identified in the DF, whereas only five are visible in the corresponding BF.

These observations are attributed to the higher sensitivity of DF imaging to small fragments and its ability to provide higher temporal resolution through the increased number of illuminations per frame.

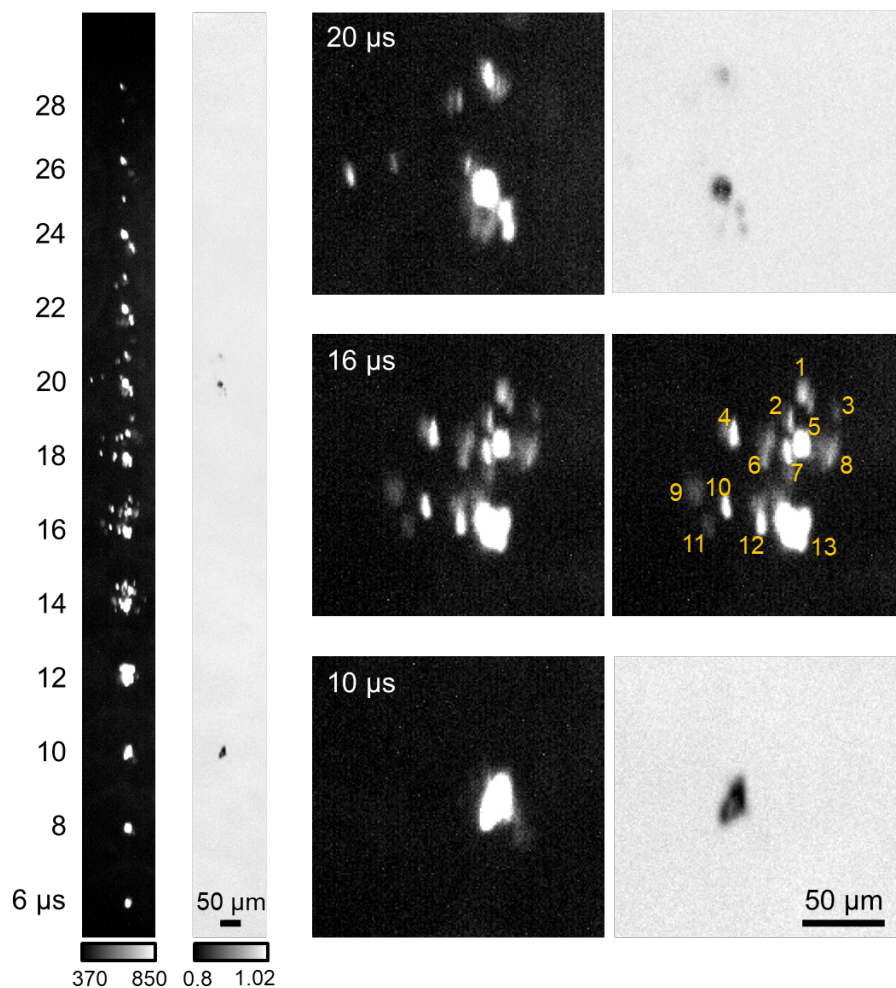


Figure 8: Dark-field and bright-field images of a disrupting droplet illuminated multiple times within the same camera frame using two synchronized cameras. The illuminations at $10\ \mu\text{s}$ and $20\ \mu\text{s}$ show corresponding dark-field and bright-field recordings. The illumination at $16\ \mu\text{s}$ shows an additional dark-field recording without a

corresponding bright-field counterpart. The higher temporal resolution and increased sensitivity of dark-field imaging allow the identification and counting of fragments formed shortly after droplet disruption, before they move out of focus and/or burn up.

Figure 9 shows a different droplet disruption event. In this case, 9 DF and one BF illuminations were recorded. Throughout the DF illumination sequence, two larger out-of-focus droplets overlap with the disrupting droplet, slightly reducing the image contrast. At $16\ \mu\text{s}$, the droplet appears to be in the process of breaking up following initial bubble growth between the illuminations at 14 and $16\ \mu\text{s}$. At $20\ \mu\text{s}$, the same number of fragments can be identified in both the DF and BF images. However, the contrast remains noticeably higher in the DF image, resulting in improved visibility of the individual fragments.

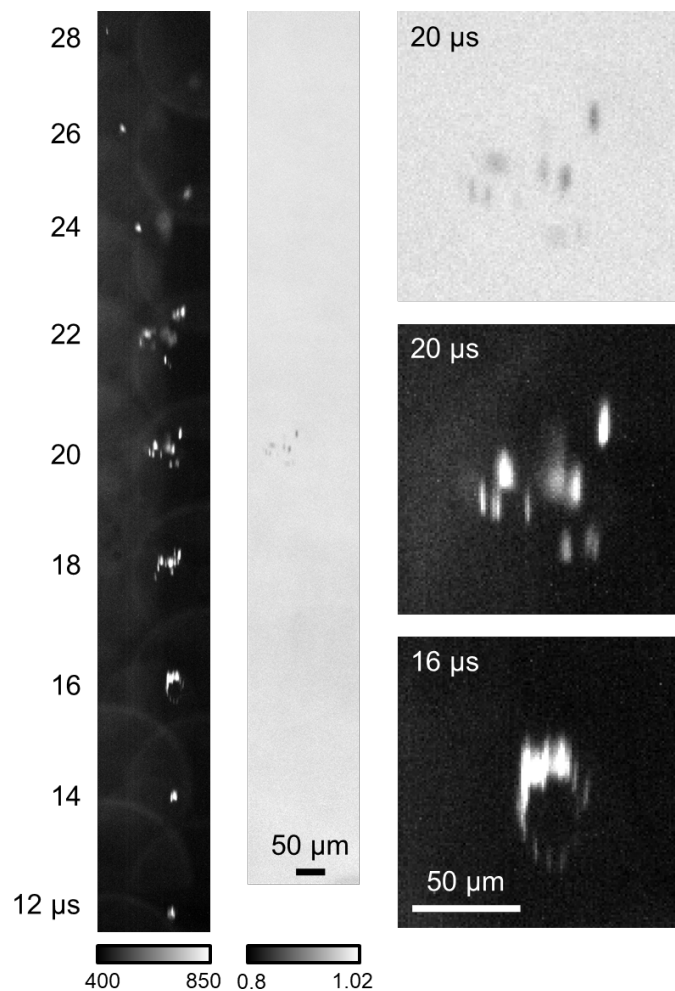


Figure 9: Dark-field and bright-field images of a disrupting droplet illuminated multiple times within the same camera frame using two synchronized cameras. The illuminations at $10\ \mu\text{s}$ show corresponding dark-field and

bright-field recordings. The illumination at 16 μs shows an additional dark-field recording without a corresponding bright-field counterpart, presumably during bubble breakup within the droplet.

During the measurement campaign, a total of 500 DF and BF frames were recorded. Of these, 47 BF frames (9.4% of all recorded frames) contained a total of 56 droplet disruption events, with up to two disruptions occurring within a single frame. The maximum number of fragments observed in a single disruption event was eight, while the average maximum visible fragment count was 3.4.

In contrast, 88 DF frames (17.6% of all recorded frames) contained a total of 131 disruption events, with up to four disruptions occurring within a single frame. The maximum number of fragments observed in a single disruption event was 14, while the average maximum visible fragment count was 5.0.

These results demonstrate a substantial increase in the number of detected droplet disruption events when using DF imaging. This finding suggests that droplet disruption may be considerably more common than previously estimated based on BF observations alone.

4. Conclusions

A combined DF and BF imaging system for the investigation of droplet disruption in spray-flame synthesis was developed and demonstrated. By exploiting the dark background of DF imaging and the velocity of the droplets, up to 15 illumination pulses could be stacked within a single camera frame, corresponding to an effective temporal resolution of 2 μs (500 kHz) between consecutive illuminations.

Comparison of DF and BF recordings showed that BF provides a more intuitive representation of the overall droplet morphology, while DF offers substantially improved sensitivity to small fragments and weakly deflecting structures. This increased sensitivity enabled the detection of a larger number of fragments during droplet disruption events and improved visibility of fragmentation processes shortly after bubble breakup.

Several limitations of the DF approach were identified. Large out-of-focus droplets may contribute to the apparent background and locally reduce image contrast. Furthermore, the large numerical apertures required for efficient DF illumination result in increased optical complexity and make precise alignment challenging.

During a measurement campaign comprising 500 image pairs, DF imaging identified 131 disruption events compared to 56 events detected in BF imaging. Furthermore, the maximum observed fragment count increased from 8 in BF to 14 in DF, while the average maximum visible

fragment count increased from 3.4 to 5.0. These results indicate that droplet disruption may occur more frequently than previously inferred from BF observations alone.

The results demonstrate that DF imaging is a promising diagnostic technique for high-temporal-resolution investigations of droplet disruption in spray-flame synthesis and other applications involving sufficiently fast-moving objects, where multiple illuminations can be stacked within a single camera frame without significant illumination overlap.

Future work should focus on increasing DF illumination intensity while reducing pulse duration to minimize motion blur and improve the signal-to-noise ratio of small fragments. Additional investigations should include quantitative fragment statistics for different precursor concentrations and a more comprehensive analysis of fragment evolution, including fragment velocities following droplet breakup.

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