

Fluid Flow Observations of the Spray Near-Field using High-Speed X-ray Imaging

Julie K. Bothell¹, Timothy B. Morgan¹, Alan L. Kastengren², and Theodore J. Heindel^{1*}

¹Center for Multiphase Flow Research and Education and Department of Mechanical Engineering, Iowa State University, USA

²X-ray Science Division, Advanced Photon Source, Argonne National Laboratory, USA

Abstract

1 Sprays are important to many industrial processes, but the near-field region where spray
2 formation takes place is difficult to visualize and characterize through visible light imaging
3 because this region is typically optically dense. X-ray imaging is one technique that can
4 penetrate this optically dense region and provide unique observations of the spray formation
5 process. This study uses high-speed white beam X-ray imaging conducted at the Advanced
6 Photon Source to capture spray formation dynamics from a canonical airblast atomizer. A range
7 of momentum flux ratios are visualized by varying the gas flow rate while the liquid flow rate is
8 held constant. The use of X-rays show overlapping flow structures including bags, ligaments,
9 mushrooms, and webs. Air bubbles are captured within the liquid regions, and a very dynamic
10 crown forms at the liquid needle exit at higher momentum flux ratios. Finally, at the highest

* Corresponding Author: Theodore J. Heindel
Department of Mechanical Engineering
Iowa State University
2043 Black Engineering
2529 Union Drive
Ames, IA 50011-2161
theindel@iastate.edu

11 momentum flux ratios, air bubbles and liquid droplets are observed migrating upstream in the
12 center of the crown, and this motion is attributed to a toroidal recirculation cell at the nozzle exit.

13 **Keywords:** Atomization, high-speed imaging, primary breakup, spray, synchrotron,
14 X-ray diagnostics

15 **1 Introduction**

16 Sprays are found in many natural and industrial settings, from rain and breaking ocean
17 waves to liquid fuel combustion and additive manufacturing. In typical applications, when liquid
18 is ejected from a nozzle in a gaseous environment and sufficient shear is available, the liquid is
19 broken up into to a liquid-gas spray. The resulting spray is commonly divided into three regions.
20 The near-field region is closest to the nozzle exit and is generally composed of a dense liquid
21 core or sheet that undergoes breakup into ligaments, which then undergo additional breakup into
22 individual liquid droplets. The spray mid-field region is usually composed of a dispersion of
23 liquid ligaments undergoing breakup and individual liquid droplets that may undergo secondary
24 breakup. Finally, the far-field region contains a dilute dispersion of individual liquid droplets that
25 continue to disperse radially and axially as they travel from the nozzle exit. While the far-field
26 region has been well characterized, primary breakup and the near-field region has been
27 minimally investigated because the thick liquid core and large structures render the region nearly
28 impenetrable to visible light. However, the near-field region is vital to understanding sprays
29 because it controls and sets the primary combustion zone for fuel sprays (Lightfoot et al., 2015).
30 Additionally, it provides the initial conditions as the spray enters the secondary breakup region.

31 The spray mid- and far-field regions can be quantified using optical- and laser-based
32 methods like shadowgraphy, back-illuminated or backlit imaging, Phase Doppler Particle
33 Analysis (PDPA), or Particle Image Velocimetry (PIV) (Bachalo, 2000; Castrejón-Pita et al.,

34 2013; Fansler and Parrish, 2015; Settles and Hargather, 2017). The challenge of using these
35 techniques in the near-field is that the spray is usually optically dense in this region; hence, other
36 techniques are required like X-ray imaging (Heindel, 2018; Kastengren and Powell, 2014;
37 Kastengren et al., 2012; Linne, 2012a, 2012b, 2013). Unlike visible light, X-rays are able to
38 penetrate the dense liquid near-field region. The X-ray attenuation (absorption) in this dense
39 region is related to the amount and density of material through which the beam passes. The
40 attenuation can be used to quantify the amount of material present in the line of sight along the
41 X-ray beam using Beer-Lambert's Law (Li et al., 2019). One additional advantage of X-rays is
42 the slight refraction and diffraction at liquid-gas interfaces, which can actually enhance
43 visualization in a technique called phase contrast imaging (Heindel, 2018; Kastengren et al.,
44 2012).

45 Traditional radiography is the act of obtaining a shadow-like image of an object using
46 penetrating radiation such as X rays or γ -rays, where darker regions correspond to where the
47 object is more dense and/or thicker (Cartz, 1995); a simple medical X-ray showing bone
48 structure is a radiograph. X-ray computed tomography is when radiographs of an object are
49 acquired from several hundred viewing directions and then reconstructed with standard
50 algorithms to show intensity variations of the object cross-section (Hampel, 2015; Hsieh, 2003).

51 X-rays have been used to visualize multiphase flows for many years (Aliseda and
52 Heindel, 2021; Heindel, 2011). They are typically generated using one of two methods: (i) an
53 electron beam hitting a metal target like that found in traditional tube sources, or (ii) a
54 synchrotron light source. The high-speed X-ray imaging in this paper was produced using the
55 synchrotron facility at the Advanced Photon Source (APS) at Argonne National Laboratory.
56 X-rays produced by a synchrotron source are polychromatic, like those produced by tube

57 sources, but they are highly collimated and produce a much higher X-ray flux (up to 6 orders of
58 magnitude higher (Matusik et al., 2019)). X-rays produced with a synchrotron source are found
59 only at selected facilities like APS (Kastengren and Powell, 2014; Kastengren et al., 2012;
60 Tekawade et al., 2020).

61 There are several X-ray imaging techniques that can be used to characterize sprays
62 (Heindel, 2018). White beam radiography or white beam imaging can be completed using a
63 synchrotron source. White beam imaging uses the high intensity X-ray beam from the
64 synchrotron that is composed of a range of energies (wavelengths), similar to white light being
65 composed of a range of wavelengths in the visible spectrum. Once the beam passes through the
66 object of interest, it is detected by a scintillator crystal that luminesces visible light proportional
67 to the amount of absorbed X-ray energy. The luminescence is imaged using a turning mirror into
68 a high-speed digital camera that is shielded from the X-rays. White beam imaging has been used
69 to investigate, for example, the liquid core dynamics of a high momentum ratio spray
70 (Machicoane et al., 2019), the two phase flow behavior in an opaque beryllium injector (Lin et
71 al., 2017), the mixing of an impinging jet spray (Halls et al., 2017), and the gas-liquid interface
72 of a turbulent liquid jet being injected into still air (Osta et al., 2012).

73 A subset of white beam imaging is called X-ray Phase Contrast Imaging (XPCI), which
74 requires a high-energy coherent X-ray beam like the white beam at a synchrotron facility. XPCI
75 is based on the differences between the refractive index of a sample and its environment. The
76 X-ray beam passes through the object of interest (i.e., fluid flow) and is allowed to propagate
77 some distance past it. A phase shift is produced at the interface of two materials with a high
78 density difference, like a gas-liquid interface, creating a Fresnel diffraction pattern that enhances
79 the absorption contrast at the interface; the enhanced absorption contrast can be used to facilitate

80 image analysis to quantify flow features. XPCI has recently been used to explore the flow
81 through a fuel injector (Moon et al., 2015), the cavitation dynamics in a high-speed nozzle
82 (Karathanassis et al., 2021), and venturi flows (Khlifa et al., 2017; Zhang et al., 2020a; Zhang et
83 al., 2020b).

84 Tomographic imaging of multiphase flows has been summarized in recent reviews
85 (Aliseda and Heindel, 2021; Heindel, 2011). This imaging modality has also been applied to
86 sprays. To acquire the necessary number of image projections, either the source and detector
87 have to revolve around the spray (Halls et al., 2014; Li et al., 2017b), the spray has to rotate
88 within the imaging region (Coletti et al., 2014; Tekawade et al., 2020), or multiple source-
89 detector pairs have to be arranged around the spray (Halls et al., 2019; Rahman et al., 2018).

90 This paper describes the flow structures from a canonical airblast atomizer that are
91 visualized using high-intensity X-rays, which allows for high-speed flow visualization (up to
92 100 kHz in this study). While the liquid flow rate is held constant, the gas flow rate is varied to
93 provide a range of momentum flux ratios, where the shear imposed on the liquid increases with
94 increasing momentum flux ratio, creating highly dynamic aperiodic flows, which are discussed
95 below.

96 **2 Experimental Setup**

97 The experimental setup for this study consists of two major components, the flow loop
98 (discussed in Section 2.1) and the X-ray imaging setup (discussed in Section 2.3). Additionally,
99 the non-dimensionalized flow parameters are defined in Section 2.2 and a brief discussion on the
100 X-ray image processing can be found in Section 2.4.

101 **2.1 Nozzle and Flow Loop**

102 The spray used in this study is generated using a canonical two-fluid airblast atomizer
103 shown in Figure 1. The inner aluminum liquid needle has an ID of $d_i = 2.1$ mm and OD of
104 $D_i = 2.7$ mm at the tip, and is coaxial with an outer air chamber ($d_g = 10$ mm ID at the tip). Two
105 identical outer air chambers are used in this work, one fabricated from aluminum and the other
106 fabricated from acetal plastic. The aluminum outer nozzle allows enough X-ray signal through to
107 view the internal flow (see, for example, Machicoane et al. (Machicoane et al., 2019) for a static
108 image or Li et al. (Li et al., 2017a) for a video). The acetal plastic nozzle has a precision copper
109 ring at the exit and is used as an electrical grounding path for a separate study involving
110 electrostatics; the copper ring prevents X-ray visualization inside the nozzle. Complete details of
111 the nozzle design are found elsewhere (Huck et al., 2018; Machicoane and Aliseda, 2017;
112 Machicoane et al., 2019).

113 Air and water are used as the two fluids in the airblast atomizer. The air and water flow
114 loops have been detailed elsewhere (Bothell et al., 2020a; Li et al., 2019) and only a summary is
115 provided here. Compressed air is provided through the building supply and is filtered prior to
116 entering the flow loop. After the filter, the air is partitioned into two lines, each of which has a
117 pressure regulator to limit the air pressure that is fed to the system. One of the compressed air
118 lines is used to feed air to the nozzle, while the second is used to pressurize a water tank to
119 provide the water to the nozzle. A ball valve on each of the air lines is used to close the air line
120 when not in use.

121 The air feed line is partitioned a second time to direct the air flow as “co-flow” and
122 “swirl” air (see Figure 1). Electronic proportioning valves and air flow meters are located after
123 this partition and are used to control and monitor the air flow rates. Each co-flow and swirl air

124 line passes through air flow meters and then each are further split into four identical lines that are
125 then attached to the upper portion of the nozzle to provide air flow into the test nozzle. The four
126 co-flow lines impinge onto the liquid needle longitudinal axis, across the four quadrants,
127 producing a purely coaxial gas stream at the nozzle exit. The four swirl lines enter the air nozzle
128 in the same plane as the co-flow lines, but tangent to the air nozzle wall, introducing swirl in the
129 gas stream. When swirl is added to the gas, the total gas flow rate remains constant, diverting
130 part of the total gas from the co-flow to the swirl injection.

131 The second air line is fed into the water tank to create a pressurized section of air on top
132 of the water that provides the force necessary to push the water through the water line. From
133 there, the water passes through a water flow meter and an electronic proportioning valve and
134 then enters the liquid needle. The liquid needle is designed to produce fully developed laminar
135 Poiseuille flow and has $L/d_l \approx 50$.

136 The electronic proportioning valves and flow meters are connected to a data acquisition
137 system that provides real time measurement and control. The electronic proportioning valves are
138 each controlled by a proportional, integral, and derivative (PID) control system implemented in
139 an in-house LabView program. Values from the flow meters are sent to the data acquisition
140 system and used as feedback, as well as stored so that any observed phenomenon can be related
141 back to the instantaneous flow measurement.

142 **2.2 Flow Parameters**

143 The total liquid (Q_l), co-flow (Q_{NS}), and swirl (Q_{SW}) flow rates are measured during each
144 test, where the total air flow rate $Q_{total} = Q_{NS} + Q_{SW}$. The ratio of swirl air flow rate to co-flow air
145 flow rate is defined as the swirl ratio (SR) to quantify the amount of air entering the gas nozzle
146 through the tangential ports relative to the amount entering perpendicular to the liquid needle:

147
$$SR = \frac{Q_{SW}}{Q_{NS}} \quad (1)$$

148 In this work, most observations are provided with $SR = 0$; a few cases are shown with $SR = 1$.

149 Note that the total gas flow rate remains constant when the swirl ratio is varied.

150 The liquid Reynolds number is defined as:

151
$$Re_l = \frac{U_l d_l}{\nu_l} \quad (2)$$

152 where U_l is the mean exit velocity of the liquid and ν_l is the kinematic viscosity of water. The gas

153 Reynolds number is defined as:

154
$$Re_g = \frac{U_g d_{eff}}{\nu_g} = \frac{U_g \sqrt{d_g^2 - D_l^2}}{\nu_g} \quad (3)$$

155 where U_g is the mean exit velocity of the gas, defined as Q_{total}/A_g with A_g the total gas exit area;

156 ν_g is the kinematic viscosity of air; and d_{eff} is the gas effective diameter at the nozzle exit,

157 defined as the diameter of a circle with the exit area equal to A_g .

158 The momentum flux ratio is defined as:

159
$$M = \frac{\rho_g U_g^2}{\rho_l U_l^2} \quad (4)$$

160 where the subscripts g and l represent the gas and liquid properties, respectively, and ρ is the

161 fluid density.

162 Finally, a Weber number for the flow can also be defined based on the mean slip velocity

163 and the liquid nozzle exit diameter:

164
$$We = \frac{\rho_g (U_g - U_l)^2 d_l}{\sigma} \quad (5)$$

165 where σ is the surface tension of water.

166 All cases investigated in this work held the liquid flow rate constant at $Re_l = 1,100$ while
167 the gas flow rate is varied such that $21,300 \leq Re_g \leq 180,000$, yielding $6 \leq M \leq 440$ and
168 $39 \leq We \leq 2,900$. This corresponds to actual mean fluid velocities at the nozzle exit of
169 $U_l = 0.48$ m/s and 34 m/s $\leq U_g \leq 290$ m/s.

170 **2.3 X-ray Imaging**

171 The high-speed flow visualization in this paper is completed over four separate
172 experimental campaigns at APS; this facility uses synchrotron technology to provide high
173 brilliance X-ray beams that can be used for flow visualization (Kastengren and Powell, 2014;
174 Kastengren et al., 2012). APS works by accelerating electrons to nearly the speed of light and
175 then bending the path of the electrons. When the electron path is bent by a bending magnet,
176 wiggler, or undulator to change their direction, they emit an X-ray beam that is tangent to the
177 direction of motion. The 7-BM beam line is used throughout this study, which provides high
178 intensity white beam X-rays that can be used for 2D radiographic imaging with microsecond or
179 sub-microsecond time resolution (Halls et al., 2017), and can produce very detailed 2D spatially
180 and temporally resolved measurements of highly dynamic 3D events, like spray formation.

181 The setup for APS white beam radiography is shown in Figure 2. A broadband X-ray
182 beam is first emitted from the synchrotron ring. It then, optionally, passes through a set of filters
183 to reduce soft X-rays and beam hardening, or to reduce the beam flux, if desired. The beam then
184 passes through a rotating chopper wheel to further reduce the time-average power of the beam.
185 The chopper wheel is composed of two 15.2 cm diameter, 6.35 mm thick copper discs with two
186 “pie wedge” openings cut into each disc. The discs are arranged such that a common open wedge
187 of about 7.9° was used. The discs are spun at 0.5 Hz, yielding bursts of X-rays at 1 Hz. Although

188 the chopper wheel is used to limit the average beam intensity, the acetal plastic nozzle still
189 suffered radiation damage at the end of the last imaging campaign (Heindel et al., 2019).

190 After passing through the chopper wheel, the beam passes through the spray where the
191 intensity of the beam decreases, depending on the amount of fluid through which it passes. The
192 beam is then detected by a scintillator crystal. In this study, either a 500 μm thick YAG:Ce or a
193 100 μm thick LuAG scintillator crystal is used, depending on the test condition. Each scintillator
194 crystal luminesces in the visible spectrum with an intensity proportional to the X-ray intensity.
195 The visible light is then reflected by a mirror into a high-speed digital camera where an image is
196 taken. The mirror is necessary because X-rays that are transmitted through the scintillator crystal
197 could damage the camera if it is in the beam path. Additionally, shielding is placed around the
198 imaging equipment to shield it from scattered X-rays that could damage the camera electronics.

199 In this study, one of two high-speed cameras are used. The first is a Photron FASTCAM
200 Mini AX50 with a frame rate of 2 kHz at full field of view (1024×1024 pixels). The second is a
201 Photron FASTCAM SA-Z with a frame rate of 20 kHz at full field of view (1024×1024 pixels).
202 In most cases, the cameras are operated using a reduce field of view to increase the frame rate.
203 The cameras are connected to 105 mm – 50 mm or 180 mm – 50 mm macro-coupled lens
204 combinations, respectively, to provide an effective magnification of 2.1x or 3.6x. At the highest
205 SA-Z frame rates (100 kHz), a 1.4x teleconverter is also added to the lens combination to yield
206 an effective magnification of 5.04x. Frame rates typically varied from 6 kHz for the slowest flow
207 rates using the AX50 to 100 kHz for the highest flow rates using the SA-Z, with exposure times
208 ranging from 1.05 μs to 3.75 μs , respectively.

209 Note the white beam in the 7-BM beam line at APS is not square (~ 8 mm wide by ~ 6 mm
210 tall), and the intensity is most uniform in the beam center and then drops off toward the top and

211 bottom edges. Furthermore, the spray is much larger than the X-ray beam and most imaging is
 212 completed using a reduced field of view to utilize a higher camera frame rate and a more uniform
 213 beam flux. Hence, the beam center was the preferred imaging location. Additionally, to capture a
 214 wider spray region, a quasi-checkerboard imaging pattern is used to image several discrete
 215 locations. As illustrated in Figure 3, a backlit image of the airblast nozzle spray shows a large
 216 spray extent while the white boxes show a quasi-checkerboard pattern used for white beam
 217 imaging of the same spray. The exact image size and checkerboard pattern depends on the
 218 specific camera, its image settings, and the area of interest within the spray. This study focuses
 219 on the near-field region and most white beam imaging is within 10 liquid needle diameters
 220 (about 2 gas nozzle diameters or 20 mm) or less from the nozzle exit. Furthermore, as the
 221 momentum flux ratio increased, the number of axial locations decreased because a full spray
 222 formed closer to the nozzle exit. Finally, the (x, y) coordinates of any given image represent the
 223 image center, where x is in the spray axial direction in mm, y is in the spanwise direction in mm,
 224 and (0, 0) represents the nozzle exit on the centerline of the liquid needle.

225 **2.4 Image Normalization and Resolvable Object Size**

226 Normalization of X-ray images helps to eliminates static bias which improves the image
 227 quality. Normalization is generally the first step in pre-processing the images for further analysis.
 228 Flat field normalization is used for this study and is defined by (Van Nieuwenhove et al., 2015):

$$229 \quad N = \frac{(P - D)}{(F - D)} \quad (6)$$

230 where P represents the original image, D is a dark field image, F is the flat field image, and N is
 231 the new normalized image. The mathematical operations in Eq. (6) are performed pixel-by-pixel.
 232 The dark field image is an image taken with no light entering the camera. The flat field image is
 233 created by taking a series of background images from a region where there is no spray or nozzle.

234 In practice, a sequence of background images (typically 400 frames) are acquired and averaged
235 together to produce a flat field image with minimal noise. Additionally, as the cameras used in
236 this study have a built-in calibration that does the dark field compensation onboard the camera,
237 an ideal image of all zeros is used for the dark image.

238 From a visual inspection of the white beam images used in this study, the minimum
239 resolvable object size is 5 pixels in diameter (20-25 μm , depending on camera and lens
240 combination). Factors that affect the minimum resolvable object size include image resolution,
241 contrast between the droplets and the background compared to the amount of noise that is
242 present, and sources of error such as distortion and blurring which can be ignored in favor of the
243 visual validation. The maximum resolvable object size is limited by the maximum size of droplet
244 that can fit within the imaging frame.

245 **3 Results and Observations**

246 To show the advantages of X-ray imaging of sprays with respect to visible light imaging,
247 Section 3.1 compares the visible features of a spray with $M = 6$ and $SR = 1$ between backlit
248 imaging and X-ray white beam imaging. Section 3.2 makes overall comparisons of how the
249 spray near-field changes as the momentum flux ratio is increased. Finally, the detailed features of
250 spray breakup and changes in the liquid core are examined in Section 3.3.

251 **3.1 Advantages of X-ray Imaging**

252 Historically, backlit or shadowgraph imaging has been a popular method for studying the
253 near-field region of a spray (Castrejón-García et al., 2011; Castrejón-Pita et al., 2013; Dash et al.,
254 2018; Settles and Hargather, 2017). However, white beam X-ray radiography has recently gained
255 traction because the technique provides information-rich images and enables rapid data
256 collection. The high intensity of the X-ray beam makes it possible to have a short exposure so

257 that fast-moving sprays can be captured without blur. The constant beam flux during image
258 acquisition also allows images to be taken with high-speed cameras, and therefore the movement
259 of the spray can be visualized (Li et al., 2017a). X-rays attenuate as they pass through liquid, so
260 the resulting image can be correlated to the amount of liquid along the beam path (Li et al.,
261 2019). Hence, the X-ray image is a 2D projection of the liquid content along the beam path,
262 where the gray scale variation correlates to liquid thickness, which provides information in the
263 depth dimension. In contrast, backlit or shadowgraphy imaging uses visible light that reflects and
264 refracts off of the gas-liquid interface and provides a binary image that shows where the
265 projected liquid is, or is not, present, but provides no information on the amount of liquid in the
266 image depth direction.

267 Figure 4 shows a comparison of a backlit image to white beam X-ray images for a spray
268 with $M = 6$ and $SR = 1$. It is important to note that backlit imaging can easily cover a larger
269 spray area than synchrotron-based white beam imaging, but tube source X-rays can be used to
270 capture large spray regions if there is a sufficient signal-to-noise ratio in the images (Heindel,
271 2018; Li et al., 2019). The major difference between backlit and white beam imaging, which is
272 important for analysis, is the level of detail that is available. Backlit imaging can provide a long
273 imaging sequence and intact length and spray angle can be easily determined from the images
274 (Bothell et al., 2020a; Bothell et al., 2020b; Morgan and Heindel, 2021). However, the resulting
275 images only show the presence of liquid (a shadow), and liquid thickness cannot be determined.
276 Additionally, if multiple fluid structures like overlapping droplets exist along the light path, they
277 appear as a single object in the recorded backlit image. In contrast, for X-ray images like those
278 shown in Figure 4, the thicker liquid corresponds to the darker region. The greyscale level can be
279 correlated to the optical depth of the liquid region (Li et al., 2019), so the darker the region, the

280 more liquid is present between the X-ray source and detector. This also allows the ability to
281 identify overlapping flow structures. The benefits of white beam X-ray images over backlit
282 images are very apparent at the nozzle exit, as shown in Figure 4b. In this region, X-rays allow
283 imaging inside the outer aluminum air nozzle. The liquid needle walls are also identified. There
284 is also clear evidence of liquid wicking up the outside of the water needle (see the large arrow
285 marked with “A” in Figure 4b), which is impossible to capture with backlit imaging. Further
286 downstream, air bubbles within the liquid region and overlapping liquid structures are also
287 visible (Figures 4c and 4d, respectively). The enhanced white regions at the air-water interface
288 are also visible in the X-ray images and are due to phase contrast effects (Heindel, 2018).

289 **3.2 Overall Spray Observations and Comparisons**

290 Figure 5 shows various white beam images at selected spray locations for three different
291 momentum flux ratios. Since the images are acquired in a checkerboard fashion (see Figure 3),
292 they are uncorrelated. For each flow condition, the image size may also differ and is a function
293 of the camera and frame rate, but the physical scale is consistent across all images to show the
294 trade-offs between frame rate and image size. Note the images presented in Figure 5 (and all
295 subsequent images) represent a typical image from the respective region, and all described
296 observations represent a compilation of observations after viewing several hundred to several
297 thousand images for each flow condition and location. Finally, the liquid flow rate is held
298 constant at $Re_l = 1,100$ for each condition, $SR = 0$, and the gas flow rate is increased to increase
299 the momentum flux ratio.

300 The top image for all momentum flux ratios shows the approximate location of the liquid
301 needle exit as the dark vertical lines, and the edge of the copper grounding ring from the acetal
302 plastic nozzle is visible at the very top of the image. Focusing initially on the $M = 6$ images, bag

303 formation is easily captured as the water and air exit the nozzle. The bags breakup into ligaments
304 as they progress downstream, which breakup into droplets. Increasing the gas flow rate such that
305 the momentum flux ratio increased by a factor of 5 ($M = 30$) and then by another factor of 5
306 ($M = 150$), shows that the extent of the intact liquid region downstream of the nozzle exit gets
307 shorter as the momentum flux ratio increases. Additionally, the flow structures that are observed
308 downstream get smaller and more dispersed. More details on these structures are provided
309 below.

310 **3.3 Qualitative Description of Spray Breakup**

311 The following is a detailed qualitative description of spray breakup for sprays with
312 various momentum flux ratios. Spray dynamic descriptors follow common identifiers, when
313 available. If a name does not exist for a particular dynamic, then identifiers are chosen to best
314 represent the given dynamic structure.

315 **3.3.1 Airblast atomizer with $M = 6$**

316 When $M = 6$ and $SR = 0$, a slowly oscillating liquid stream is observed right at the liquid
317 nozzle exit. Some of the water wicks up the outside of the liquid nozzle (Heindel, 2018); X-rays
318 allow for this observation because they penetrate the opaque nozzle. As the spray progresses
319 downstream, instabilities form along the gas-liquid interface where the gas tries to penetrate the
320 liquid, forming bags as shown in Figure 5 at $M = 6$. The bag grows and deforms the liquid jet
321 leading to ligament formation, shown in Figure 6a as structures identified as “A”. As the bag
322 thins, ligaments form that are connected by a thinning liquid sheet. As the liquid moves further
323 downstream, the liquid sheet continues to thin and eventually ruptures, shown in Figure 6a as
324 structures identified as “B”. Figure 6b has been enlarged to emphasize the edge of a liquid sheet
325 (“B”) right before rupture. When bag breakup occurs, ligaments and many small droplets (with

326 respect to the size of the ligaments) form. The thinnest portion of the bag produces the smallest
327 droplets whereas the thicker edge of the bag generates the ligaments and larger droplets.

328 The liquid ligaments eventually separate from the main liquid stream at a distance known
329 as the intact length (where the continuous liquid region is no longer observed). The intact length
330 is not a constant value, but has sharp oscillations where a ligament may form at the tip and then
331 abruptly break off. Because white beam imaging of the spray is completed in sections, and the
332 intact length may span more than one image region, it is challenging to provide a precise
333 measure with the imaging technique employed here. A comparison of intact length measures
334 with different techniques has been completed for the sprays shown here (Bothell et al., 2020a).

335 As ligaments continue downstream, they can be stretched by the high-speed air
336 surrounding them, increasing the ligament speed with downstream distance. This also leads to
337 secondary bag and ligament formation, as shown in Figure 6c with the structures identified as
338 “C”; these structures continually stretch, become thinner, and eventually breakup into smaller
339 droplets.

340 The separated ligaments stretch as they move downstream, but the process is not uniform.
341 For example, Figure 7 shows an image sequence of ligament stretching and breakup, where the
342 time between frames is 555 μs (several frames are omitted between each image). Bulges in the
343 ligament “A” are connected by thin liquid regions. The bulges grow and the connecting regions
344 thin and eventually rupture, as shown by “B” and “C”, where the bulges form droplets, identified
345 by “D”. The breakup process forms nonuniform droplets as well as many satellite droplets, as
346 shown by the enhanced image region and objects identified as “E”.

347 The motion of the ligaments is quite dynamic, with different parts of the ligament moving
348 at different velocities. They can stretch axially, collapse axially to form mushroom shaped

349 structures (Fig. 8a), or tumble. Horizontal ligaments generally stretch more in the middle to
350 create a U-shape and then break apart. These structures may rupture further along different paths,
351 depending on if they are large or small. The larger mushroom structures often split into
352 secondary ligaments, or the upper and lower portions separate into two droplets. Smaller
353 mushroom structures often form a bag before breaking into smaller droplets.

354 A unique phenomenon that is visible using X-ray imaging, but not shadowgraphy or
355 backlit imaging, is air bubbles trapped inside liquid regions. When bubbles form because of bags
356 folding on the edge of the liquid stream, they move significantly slower than the rest of the
357 liquid, and some have been observed to migrate upwards towards the nozzle exit. Larger bubbles
358 may disappear as the liquid separates into ligaments and droplets. Smaller bubbles often get
359 trapped inside the liquid and move with it. Examples of air bubbles inside the liquid are
360 highlighted in Figure 8, where they are found in the observed mushroom structure, inside a liquid
361 ligament, and inside individual droplets. Bubbles trapped inside liquid droplets could have a
362 significant effect on the combustion process if the spray was a fuel injector. Bubbles could also
363 create hollow metal powders from liquid metal sprays that could be detrimental to additive
364 manufacturing processes (Anderson et al., 2018).

365 In addition to breakup, the liquid structures (ligaments, bags, mushrooms, liquid sheets,
366 and droplets) sometimes collide with each other. During collisions, the masses often combine;
367 they either remain as a single droplet or may undergo secondary breakup downstream. A series
368 of images showing droplet collision is provided in Figure 9, where several image frames are
369 omitted from this sequence to provide a representation of the collision process.

370 Similar spray structures continued to form and breakup throughout the near-field region
371 until the spray is broken into many droplets. Once the spray is composed of only droplets, the

372 droplet size distribution begins to stabilize. Secondary atomization, with droplets breaking apart
373 into smaller droplets, may still occur, but at a slower rate than primary atomization. Droplets that
374 are most likely to break up during secondary atomization are typically larger and the breakup is
375 caused by instabilities that form on the droplet surface. When secondary breakup occurs, the
376 droplets first deform into a thin oblong shape. The center region of the oblong shape narrows as
377 the ends are pulled apart. Once the center of the oblong shape becomes thin enough, the droplet
378 will suddenly break apart into two droplets, usually of similar size. The droplets move with
379 approximately the same downstream velocity but continue moving further away from each other
380 after breakup. One or more satellite droplets may also form between the larger separated
381 droplets, where the satellite droplets are much smaller than the two main droplets. A typical
382 example of this process is shown in Figure 10, where the secondary breakup of two droplets are
383 observed simultaneously. Note the time between each image is 1.1 ms.

384 3.3.2 Airblast atomizer with $M = 30$

385 Many of the phenomena that are observed from the airblast atomizer at $M = 6$ are also
386 captured when $M = 30$, but the liquid breakup occurs much faster and closer to the nozzle exit,
387 which results in smaller downstream droplets. With a more active spray region, some of the
388 breakup mechanics are too fast to capture in sequence unless the frame rate is greater than
389 45 kHz. The higher momentum flux ratio also results in more interference with overlapping
390 spray dynamics.

391 When the momentum flux ratio increases to $M = 30$ and $SR = 0$, wicking around the
392 nozzle is not visible from white beam X-ray radiographs as it was with $M = 6$. Necking occurs in
393 the attached liquid portion of the spray very near the liquid nozzle exit, identified as “A” in
394 Figure 11a with the narrowest point $0.5-1.0d_i$ downstream of the exit plane. Ligament formation

395 and eventual separation is observed after the narrowing liquid region, identified as “B” in Figure
396 11a. The separation and breakup of ligaments is rapid in comparison to a spray with $M = 6$, and a
397 larger number of liquid sheets and bags are present in the spray region just below the liquid
398 necking.

399 In sequential images in regions like that of Figure 11a, the majority of the mass appears
400 to oscillate back and forth across the 2D image projection. It is hypothesized that the oscillation
401 is more of a 3D helical flow with the direction varying in a chaotic manner. Stereographic
402 imaging is needed to confirm this. However, manual tracking of the liquid motion shows that the
403 motion did not follow a sinusoidal pattern. In comparison to the spray with $M = 6$, a spray with
404 $M = 30$ has ligaments that are much narrower, but still create liquid sheets between the ligaments
405 before they quickly break apart. With the increased shear forces from the higher gas flow rate,
406 the ligaments become very thin so that when they do break, much smaller droplets are observed
407 compared to those at $M = 6$.

408 At the higher gas flow rate, liquid sheets form at a high rate where some rejoin with
409 liquid ligaments and others become bags that break apart into small droplets. The droplets that
410 form from the bag rim are larger than those that form from the bag, but they are still smaller than
411 those observed at $M = 6$. After initial ligament formation, the liquid structures transition from
412 individual ligaments to ensembles that appear web-like, as shown in Figure 11b. The webs
413 contain ligaments, identified as “C” and ligaments with bulges and necking, identified as “D”.
414 Many of the ligaments are connected by liquid sheets, identified as “E”. These web-like
415 structures are highly dynamic, and as they travel downstream they start to breakup. Breakup
416 begins with the thin liquid sheet regions, which create the smallest droplets, and then progress to
417 ligament breakup. This process continues until the spray is composed entirely of droplets. Some

418 of the droplets contain bubbles (Figure 11c), and some form mushroom-like structures (Figure
419 11d). In both cases, these structures are smaller than those observed with $M = 6$.

420 Figure 12 provides a series of images separated by $278 \mu\text{s}$ showing the expansion and
421 breakup of a web-like structure. Note the primary shapes in each image change from frame-to-
422 frame due to the shear forces imparted on the liquid from the gas stream. Many of the larger
423 structures in Figure 12 elongate and undergo secondary breakup. When secondary breakup
424 occurs at $M = 30$, the process starts in a similar fashion to that observed with $M = 6$, where
425 droplets first deform into a thin oblong shape. The center region of the oblong shape then
426 narrows as the edges pull apart. It is common to see the oblong shape get stretched into a thin
427 ligament and break into multiple droplets if enough liquid mass is present. It is also common to
428 see a small bag form near the center of the oblong shape, which abruptly disappears upon
429 secondary breakup. A series of images, separated by $278 \mu\text{s}$, showing an oblong object forming a
430 bag and then breaking up is shown in Fig. 13. Note, this bag formation process is similar to that
431 observed with $M = 6$, but the bags and resulting droplets are much smaller at the higher gas flow
432 rate with $M = 30$.

433 The majority of the liquid spray mass continues to sweep across the spray field even
434 when the spray is all droplets. Backlit visible light imaging at this condition shows that this
435 sweeping motion is due to the upstream oscillatory motion, where the liquid continues to move
436 in the direction that is observed upstream.

437 3.3.3 Airblast atomizer with $M > 30$

438 The spray dynamics are similar when $30 \leq M \leq 66$, but change considerably when
439 $M = 95$ ($66 < M < 95$ were not visualized). Most of the formation phenomena occur very near
440 the nozzle exit. Figure 14 shows sample images at the nozzle exit for various gas flow rates

441 while $SR = 0$. All of these images were acquired with the SA-Z camera and the specific camera
442 frame rates and image sizes are summarized in Table 1. Note the physical scale of each image is
443 identical. The image (x, y) coordinate that identifies the image center relative to the liquid needle
444 exit is also provided. Additionally, videos for each of these conditions can be found in the
445 supplemental material (for paper production team: Please add link to supplemental material,
446 which have been uploaded to the journal submission web site).

447 Recall that the liquid needle has an ID of $d_1 = 2.1$ mm ID and an OD of $D_1 = 2.7$ mm; this
448 is represented in Figure 14 with the black bars on top of each image. When $M = 6$, the liquid
449 intact length extends beyond the image region. Bags also continuously form and extend beyond
450 the image region. The liquid has been observed to wick up the outer diameter of the liquid needle
451 so as it exits nozzle, the intact liquid core extends spanwise beyond the inner wall. This has also
452 been shown numerically if the wetting dynamics are properly modeled (Vu et al., 2021).

453 Increasing the momentum flux ratio to $M = 30$ causes the liquid region to narrow as it
454 exits the liquid needle and appear to oscillate in this 2D projection. Bags that do form for this
455 flow condition are short-lived and breakup into ligaments that appear to be pulled from the liquid
456 region. Air bubbles are also observed to form in the liquid region of the image, and some are
457 transported into the next image region while others disappear as they pop.

458 Similar dynamics are observed when $M = 48$ and $M = 66$. The main difference is the
459 intact length decreases with increasing M . When $M = 95$, the flow structure appears to form an
460 upside-down crown, where the inside of the crown is a depressed region of low liquid content. It
461 is hypothesized that as high velocity gas exits the nozzle around the periphery of the slower
462 liquid region, a toroidal recirculation cell forms with high velocity gas pushing the liquid
463 upstream in the center of the liquid needle. This results in liquid being pulled from the outer

464 region and a liquid depression forms in the inner region, creating the crown. Machicoane et al.
465 (Machicoane et al., 2019) have shown that when swirl is added to this flow, the crown formation
466 occurs at lower momentum flux ratios.

467 Increasing the momentum flux ratio to $M = 150$ reveals the crown walls periodically get
468 very short such that a portion may disappear. Using the aluminum outer air nozzle, Machicoane
469 et al. (Machicoane et al., 2019) showed that when this happens, the crown partially dewets from
470 a portion of the liquid needle exit, and this process is very dynamic; they called this flow
471 “unstable crown”. Air bubbles also periodically form in the inner region of the crown and then
472 dissipate.

473 Further increasing the gas flow rate to $M = 210$ shortens the crown region and it still
474 remains unstable. Some of the liquid ligaments that are pulled from the crown edges are caught
475 in the recirculation cells and wrap upstream as they dissipate. Air bubbles also appear to migrate
476 upstream in the liquid flow. At the highest momentum flux ratio of $M = 440$, the crown is very
477 short and still unstable. One interesting observation at this momentum flux ratio is that some of
478 the small liquid droplets that do form migrate upstream of the flow direction in the nozzle center.
479 We believe this happens in the center region of the liquid needle and not outside the liquid
480 needle because the high velocity gas stream outside the liquid region would prevent liquid
481 backflow. This confirms the presence of a toroidal recirculation cell where high velocity gas is
482 pushing the liquid in the center of the flow in the opposite direction.

483 The upstream migration of air bubbles and liquid droplets is further confirmed in
484 Figure 15, where images taken at 100 kHz are shown right at the nozzle exit for $M = 210$ and
485 $M = 440$ with $SR = 0$. Note that for this image acquisition speed, the image size, which is shown
486 on the figure, is reduced to a region smaller than the liquid needle ID and each image is centered

487 on the liquid needle centerline. Each image in the shown sequence is separated by 50 μs (every
 488 5th image at a frame rate of 100 kHz). When $M = 210$, the air bubble caught in the interior of the
 489 liquid crown migrates upstream over the 200 μs time frame, as shown by the white circled
 490 region. When $M = 440$, a liquid droplet is identified in the white circled region and it also
 491 migrates upstream over the given time period. It is unlikely that observations such as those
 492 shown in Figures 14 and 15 could be made with backlit or shadowgraph imaging.

493 **4 Future Applications and Challenges**

494 High-speed white beam imaging is a powerful flow visualization technique that can be
 495 used to capture high-speed multiphase flows that are obscured when using visible light or lasers.
 496 The current work shows qualitative observations. Quantitative measures of equivalent path
 497 length (EPL), defined as the accumulated liquid along the beam path, are easily acquired using
 498 focused beam measurements (Bothell et al., 2020a; Kastengren et al., 2014; Li et al., 2019). In
 499 this imaging mode, Beer-Lambert's Law can be used to determine EPL:

$$500 \quad \text{EPL} = \ell = \frac{1}{\mu} \ln \left[\frac{I_0}{I(t)} \right] \quad (7)$$

501 where μ is the attenuation coefficient. In white beam imaging, however, μ is not known precisely
 502 because it is a function of X-ray wavelength, and white beam imaging has a variety of
 503 wavelengths, which are differentially attenuated, depending on the material and material
 504 thickness. This complicates the analysis, but with proper calibration and X-ray energy modeling,
 505 the white beam-determined EPL may be approximated; this is the focus of some of our future
 506 efforts.

507 Additionally, quantitative image analysis to determine droplet size and velocity will be
 508 completed in the future. Challenges remain, however, in that multiple droplets imaged with

509 X-rays can be easily tracked with the human eye, even when they overlap for a period of time.
510 Developing imaging analysis tracking software that reliably captures these droplets is more
511 complicated due to overlapping structures and small variations in X-ray image contrast.

512 Another challenge is that the X-ray beam from the APS synchrotron facility is limited in
513 size, so the maximum field of view is only ~8 mm wide by ~6 mm tall. Additionally, the beam
514 flux near the edges of the beam are not as uniform or steady as near the center, and these
515 fluctuations will create X-ray image contrast variations with time and location.

516 Finally, the X-ray beams generated at APS are fixed in space and correspond to tangents
517 of the synchrotron ring. Hence, X-ray flow visualization from multiple projections at the same
518 instant in time, like those from two or more tube X-ray sources (Halls et al., 2019; Heindel et al.,
519 2008; Mudde, 2010), is extremely challenging. One group has attempted this (Hoshino et al.,
520 2011), but their synchrotron beam width was very large (~300 mm), and only two projections
521 were acquired simultaneously with an angle between the projections of only about 15°.

522 One possible method to do computed tomography of sprays using white beam imaging is
523 to mount the spray on a rotation system, and then rotate the spray to produce the multiple views
524 required for tomographic reconstruction. If the spray is periodic like a liquid fuel injector, it is
525 possible to phase-lock the measurements to produce time-resolved 3D images, assuming the
526 injection process is highly repeatable (Tekawade et al., 2020). Additionally, to view multiple
527 spray locations for larger sprays like the one used in this study, the spray has to be mounted on
528 an x-y translation stage. The desire to look at pressurized spray systems requires a small
529 pressurized spray facility where the entire system can be mounted on an x-y translation stage
530 (Kastengren et al., 2009), or the entire spray nozzle and x-y translation stage has to be mounted
531 within a pressured facility (Burtnett et al., 2021).

532 **5 Conclusions**

533 This observational study showed that high-speed white beam X-ray imaging, using the
534 Advanced Photon Source at Argonne National Laboratory, provided unique advantages over
535 traditional optical high-speed imaging techniques like backlit imaging. The increased detail and
536 high temporal and spatial resolution enabled a very detailed qualitative analysis of the spray
537 structure, dynamics, and breakup. Using a canonical airblast atomizer, overlapping bags
538 ligaments, and bubbles were clearly observed with X-ray imaging. As the momentum flux ratio
539 increased with a constant liquid flow rate, the atomization process moved toward the nozzle exit.
540 Bag and ligament formation still occurred, but at a faster rate. When $M \geq 95$, an unstable crown
541 formed at the liquid needle exit, and as the momentum flux ratio increased, the crown became
542 shorter and more dynamic.

543 One unique advantage of high-speed white beam X-ray imaging is that overlapping flow
544 structures can be captured including ligaments, droplets, mushrooms, and webs. Furthermore, air
545 bubbles within liquid regions were clearly captured. This also allowed for the observation that air
546 bubbles and liquid droplets migrated upstream within the liquid crown at the highest momentum
547 flux ratios on this study. These structures are not observed with optical imaging techniques.

548 One challenge with high-speed white beam X-ray imaging is the limited field of view. To
549 capture the entire spray region, particularly at the lower momentum flux ratios, several
550 overlapping imaging regions were required, and some of the flow structures could not be tracked
551 from region to region.

552 **6 Acknowledgements**

553 This work was sponsored by the Office of Naval Research (ONR) as part of the
554 Multidisciplinary University Research Initiatives (MURI) Program, under grant number N00014-

555 16-1-2617. The Photron SA-Z camera was purchased as part of the ONR DURIP program, under
556 grant number N00014-18-1-2380. The views and conclusions contained herein are those of the
557 authors only and should not be interpreted as representing those of ONR, the U.S. Navy, or the
558 U.S. Government.

559 This work was performed at the 7-BM beamline of the Advanced Photon Source, a U.S.
560 Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of
561 Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. The use of
562 their facilities and personnel is greatly appreciated.

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715 of Internal Two-Phase Flow Structures and Dynamics of Quasi-Stable Sheet Cavitation by
716 Fast Synchrotron X-ray Imaging, *Physics of Fluids*, vol. **32**, no. 11, pp. 113310, 2020b.
717

718 **Tables**

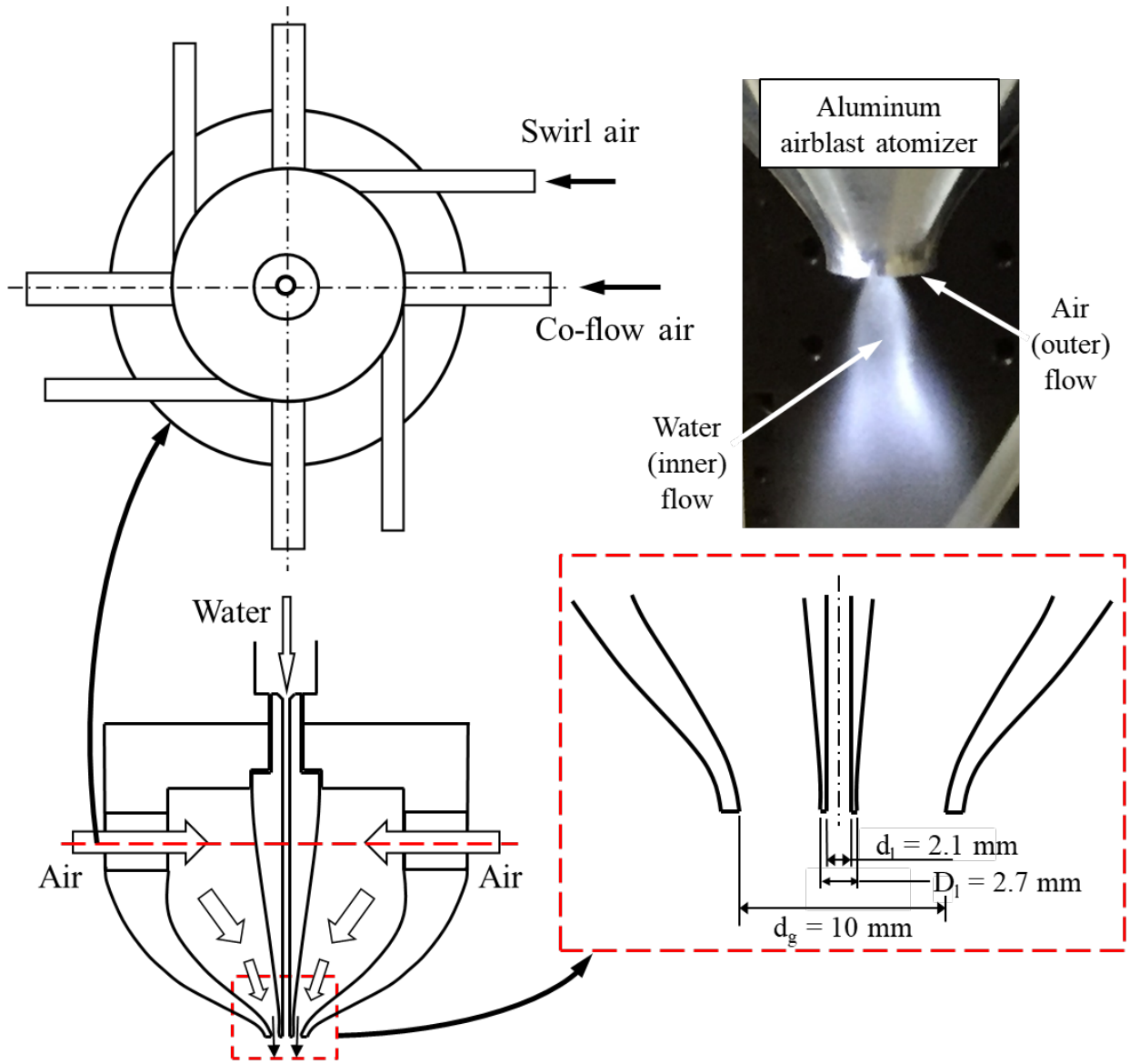
719 Table 1: Gas flow rates and camera settings for the nozzle exit images shown in Figure 14.

720 The liquid flow rate was fixed for all conditions at $Re_l = 1,100$ ($Q_l = 0.099$ LPM) and721 $SR = 0$.

Q_g (SLPM)	Re_g	M	Frame rate (kHz)	Frame size, H × V (pixels)	Frame size, H × V (mm)
150	21,300	6	36	896 × 640	4.82 × 3.56
330	46,700	30	36	896 × 640	4.82 × 3.56
420	59,600	48	45	640 × 664	3.56 × 3.69
490	69,600	66	45	640 × 664	3.56 × 3.69
590	83,800	95	45	640 × 664	3.56 × 3.69
730	104,000	150	67.2	512 × 520	2.84 × 2.89
870	124,000	210	67.2	512 × 520	2.84 × 2.89
1270	180,000	440	60	768 × 424	3.05 × 1.68

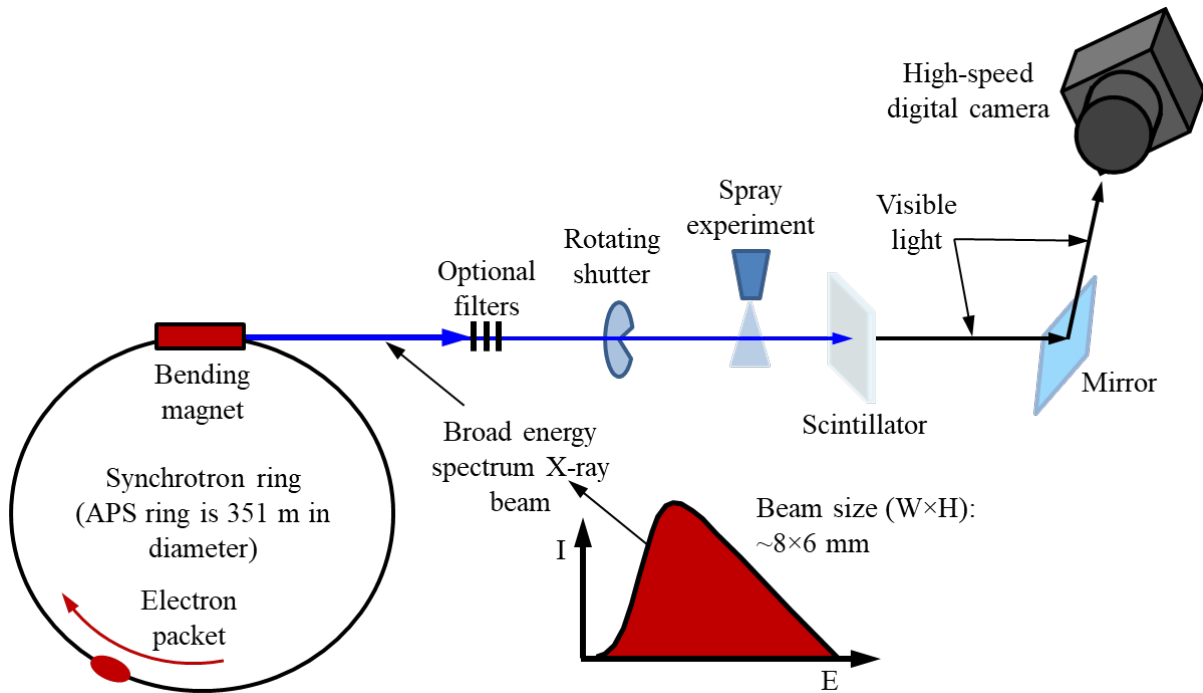
722

723 **Figures**



724

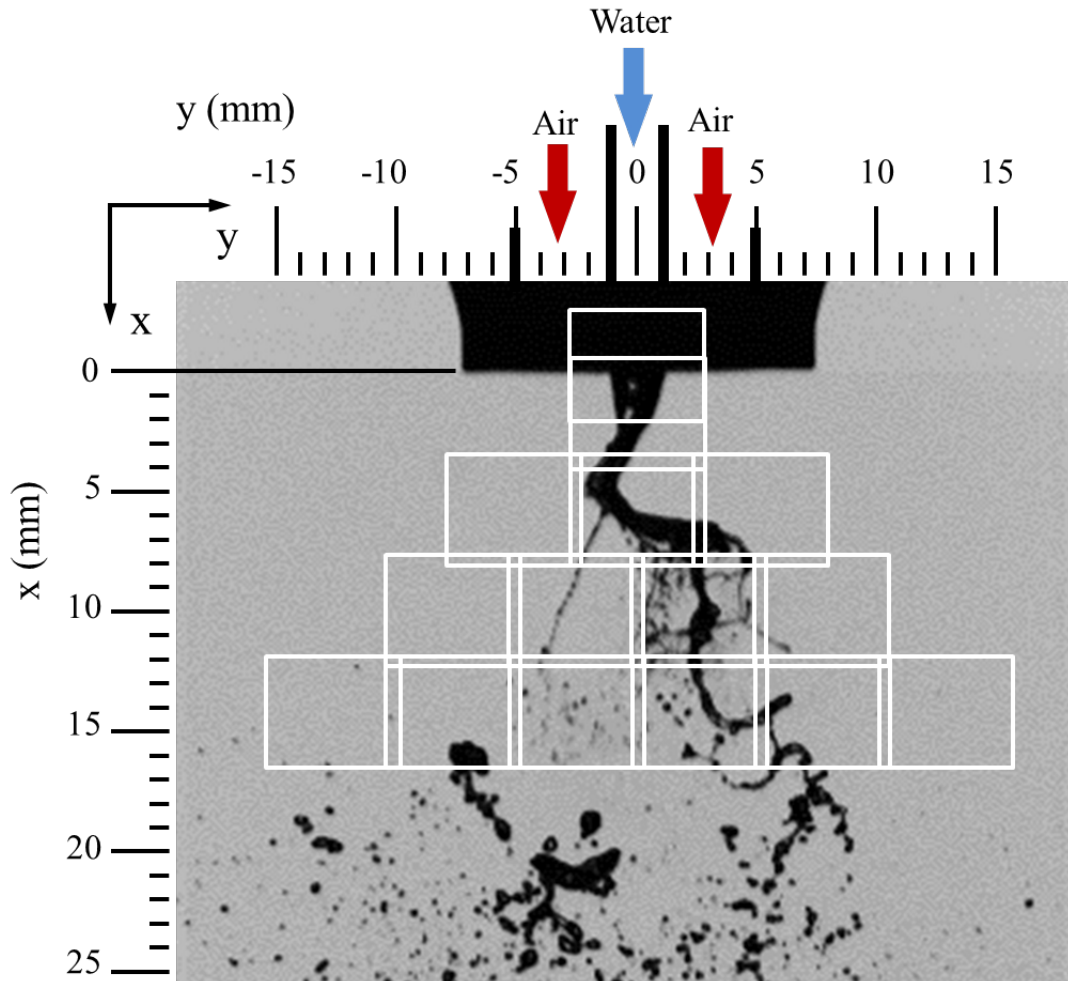
725 Figure 1: Airblast atomizer used in this study.



726

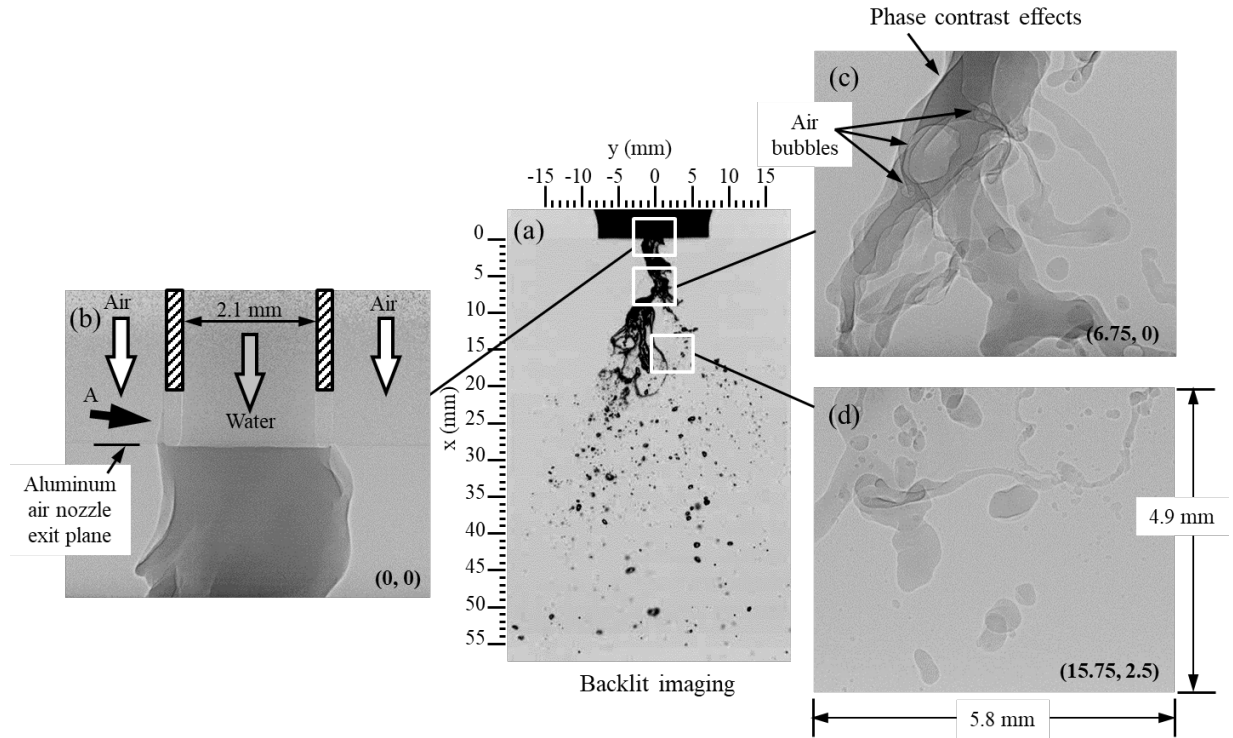
727 Figure 2: Schematic of the Advanced Photon Source (APS) experimental setup for high-speed

728 white beam imaging (not to scale).



729

730 Figure 3: Sample of overlapping image regions for white beam radiography (white boxes)
731 overlaid on a backlit image of a representative spray with $M = 6$ and $SR = 0$ (units
732 in mm).



733

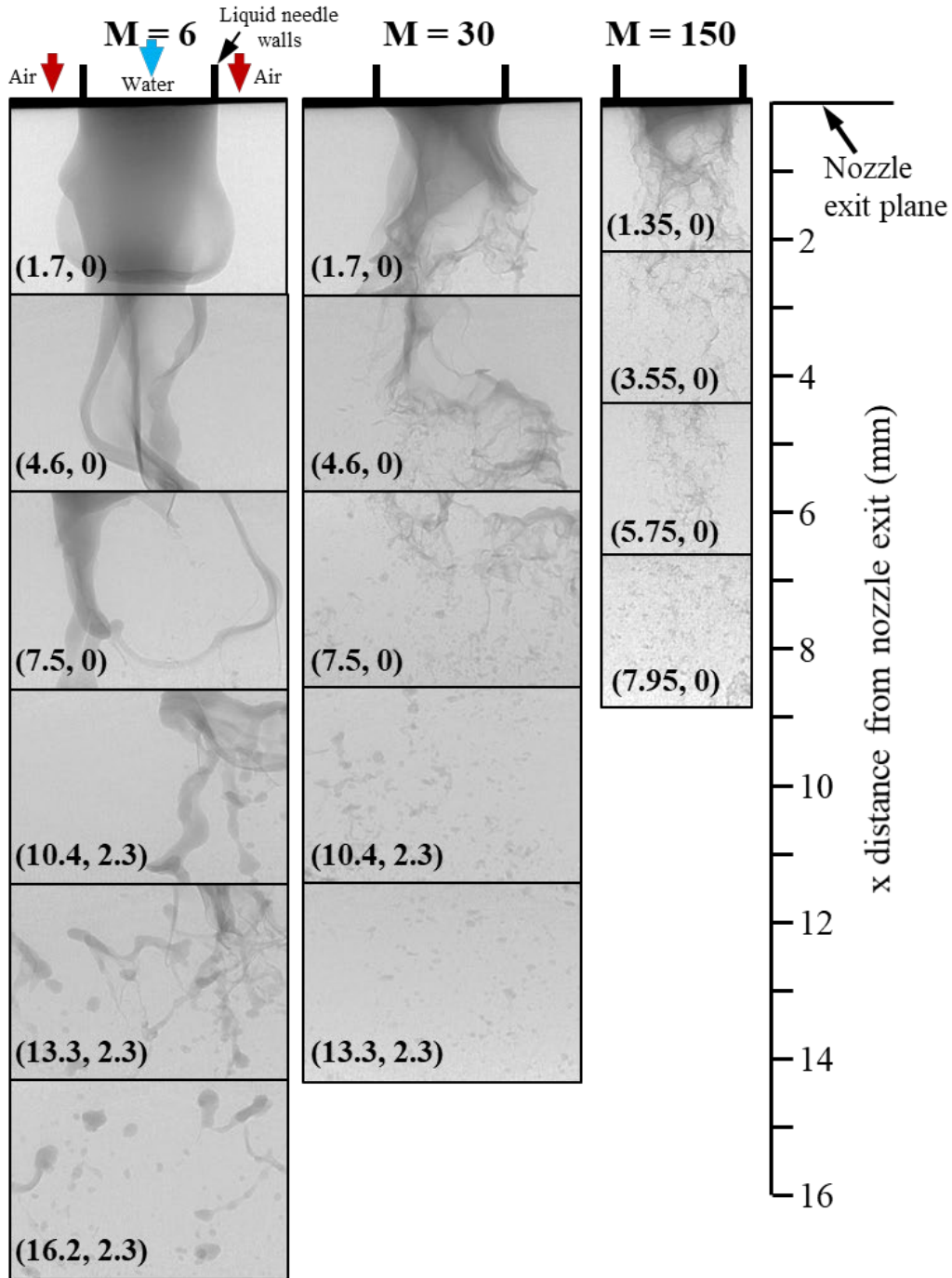
734 Figure 4: Co-axial spray with $M = 6$ and $SR = 1$. (a) Backlit image of large spray region.

735 (b) White beam X-ray image at the nozzle exit, $(0, 0)$. (c) White beam X-ray image

736 downstream of the nozzle exit, $(6.75, 0)$. (b) White beam X-ray image further

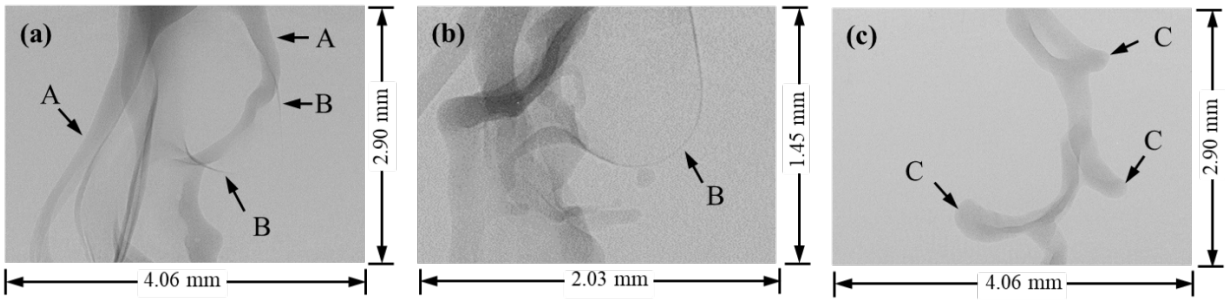
737 downstream, $(15.75, 2.5)$. All coordinate locations correspond to the (x, y) image

738 center and are given in mm.



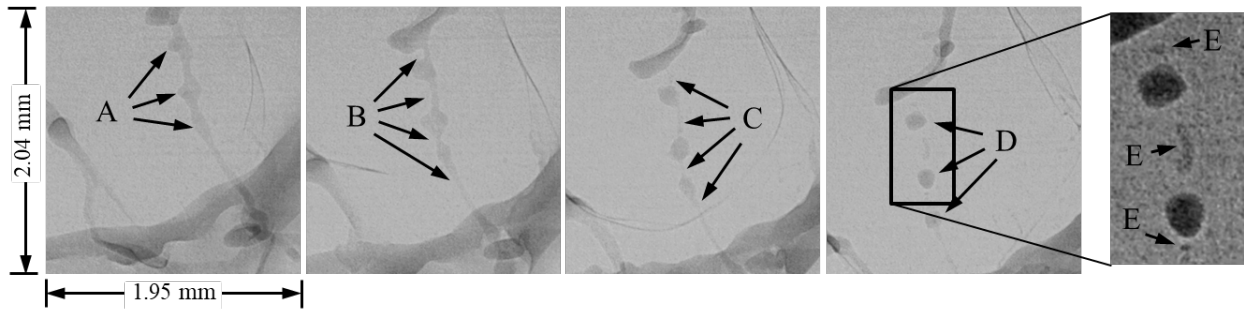
739

740 Figure 5: White beam X-ray images of an airblast atomizer showing how the spray changes in
 741 response to increasing the momentum flux ratio and downstream distance. Coordinate
 742 locations correspond to the (x, y) image center and are given in mm. Note lower
 743 images are slightly off-center.



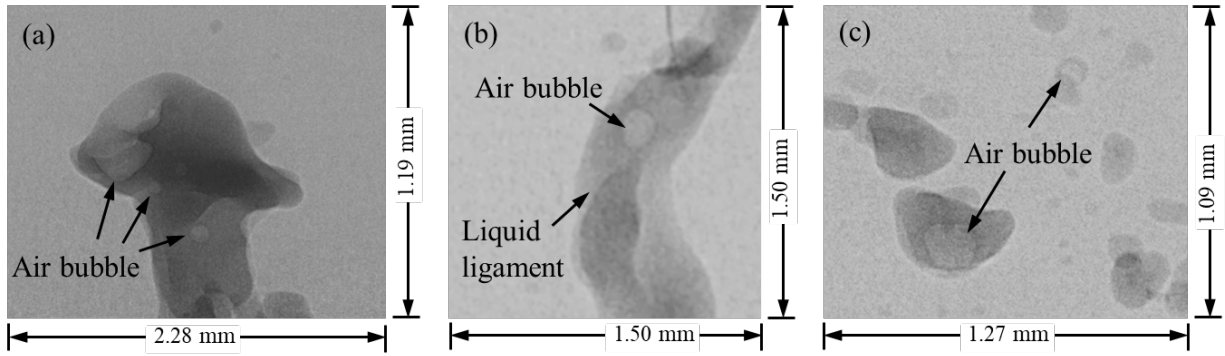
744

745 Figure 6: Selected spray structures observed with white beam X-ray imaging from an airblast
 746 atomizer with $M = 6$ and $SR = 0$. (a) Initial ligament formation; (b) bags, ligaments,
 747 and thin liquid sheets (note the image size has been increased for emphasis); and (c)
 748 secondary bag and ligament formation.



749

750 Figure 7: Ligament breakup at $M = 6$ with time between frames of $555 \mu s$. Structures identified
 751 as bulges (A), thin liquid regions connecting the bulges (B and C), thin regions
 752 rupture forming droplets (D) and satellite droplets (E).



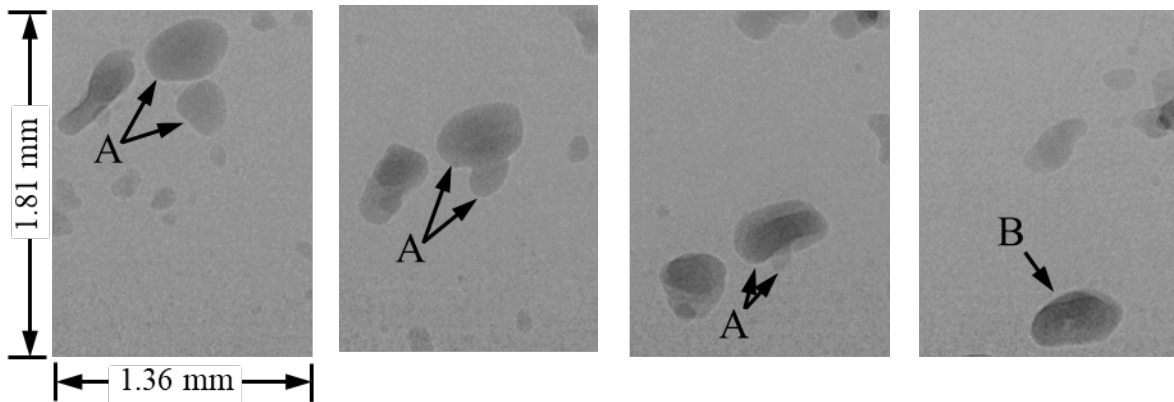
753

754 Figure 8: Sample spray structures recorded with white beam X-ray imaging with $M = 6$: (a) a

755 mushroom-like structure, (b) an air bubble inside a liquid ligament, and (c) air

756 bubbles inside liquid droplets. Note the image size varies to emphasize the given

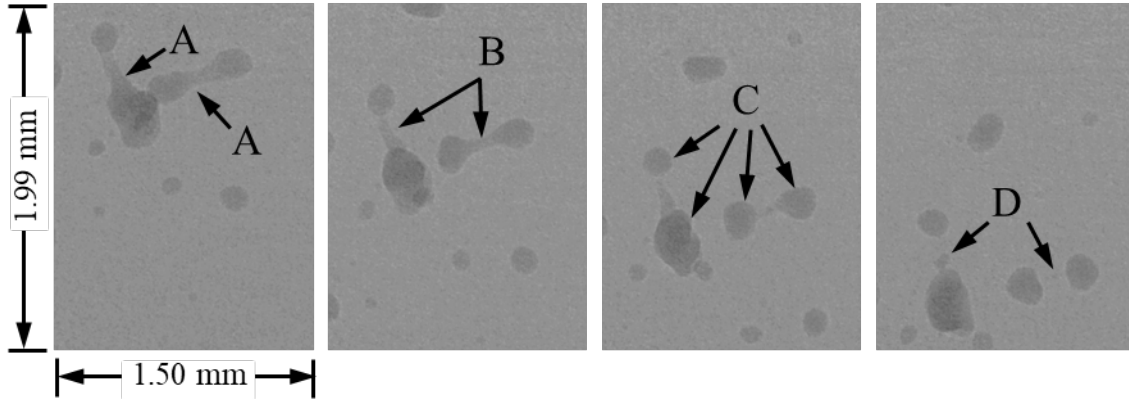
757 structure.



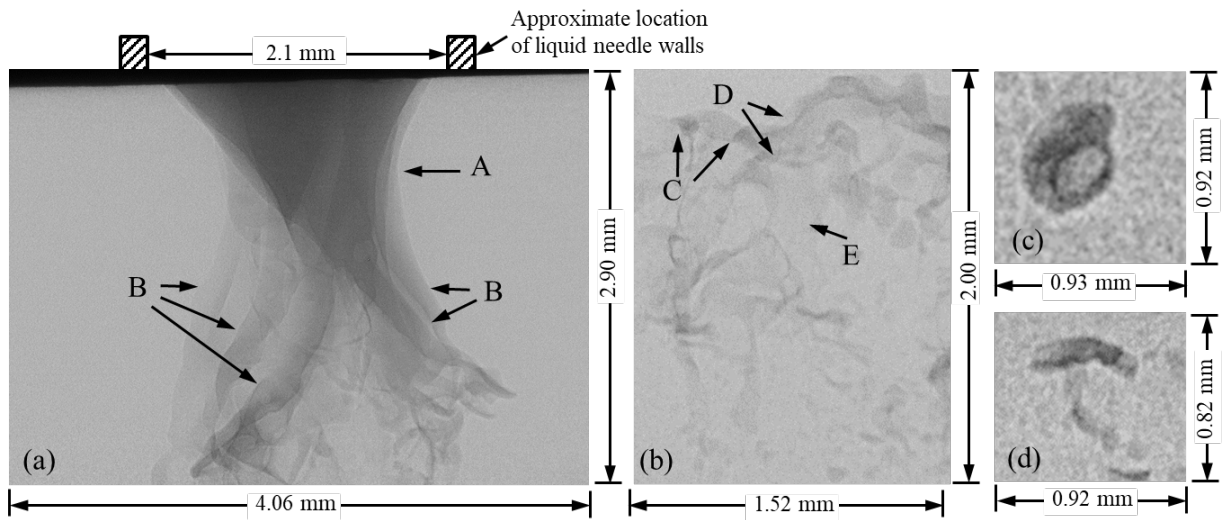
758

759 Figure 9: Droplet collision at $M = 6$ with time between frames of 1.1 ms. The two original

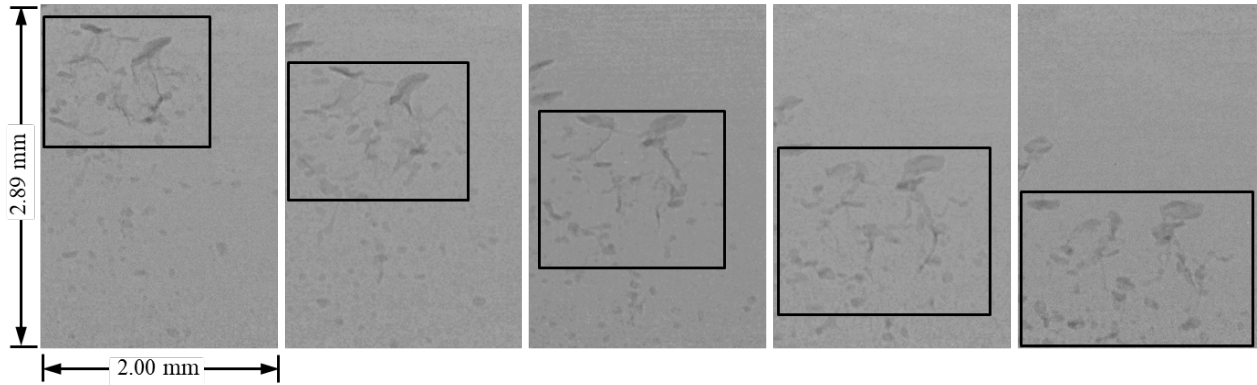
760 droplets (A) and the coalesced droplet (B).



761
 762 Figure 10: Secondary droplet breakup with $M = 6$. Oblong structures (A) are first elongated and
 763 stretched (B). The elongated droplets break and form secondary droplets (C) and
 764 satellite droplets (D).



765
 766 Figure 11: Spray formations seen in white beam X-ray images from a coaxial spray with $M = 30$
 767 and $SR = 0$. (a) Liquid necking (A) and ligament formation (B); (b) liquid web
 768 structure containing liquid ligaments (C), ligament bulges and necking (D), and liquid
 769 sheets connecting the ligaments (E); (c) bubble in a liquid droplet; and (d) mushroom-
 770 like structure.

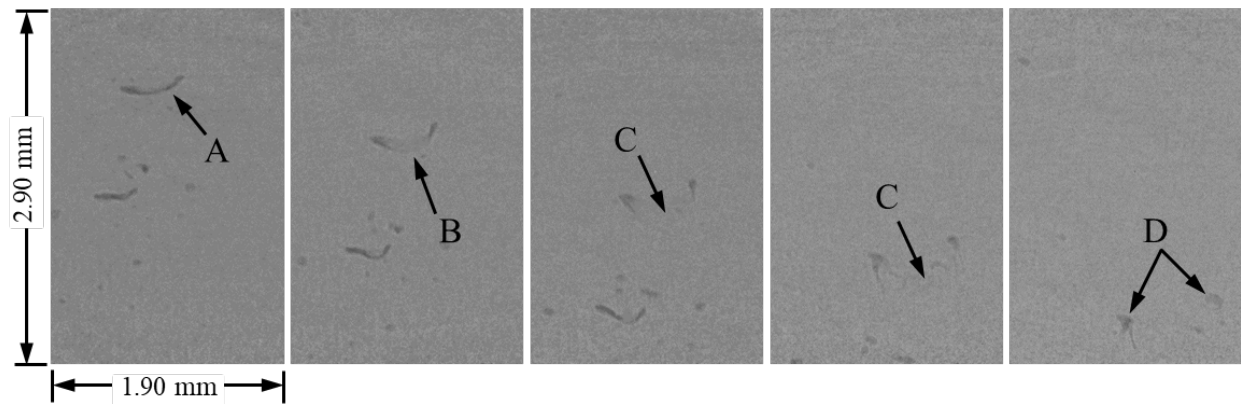


771

772 Figure 12: Downstream spray web-like structures observed with white beam X-ray imaging and

773 $M = 30$. Each image is separated by $278 \mu\text{s}$. The black box tracks a web-like structure

774 as it moves through the spray field, expanding and breaking apart.

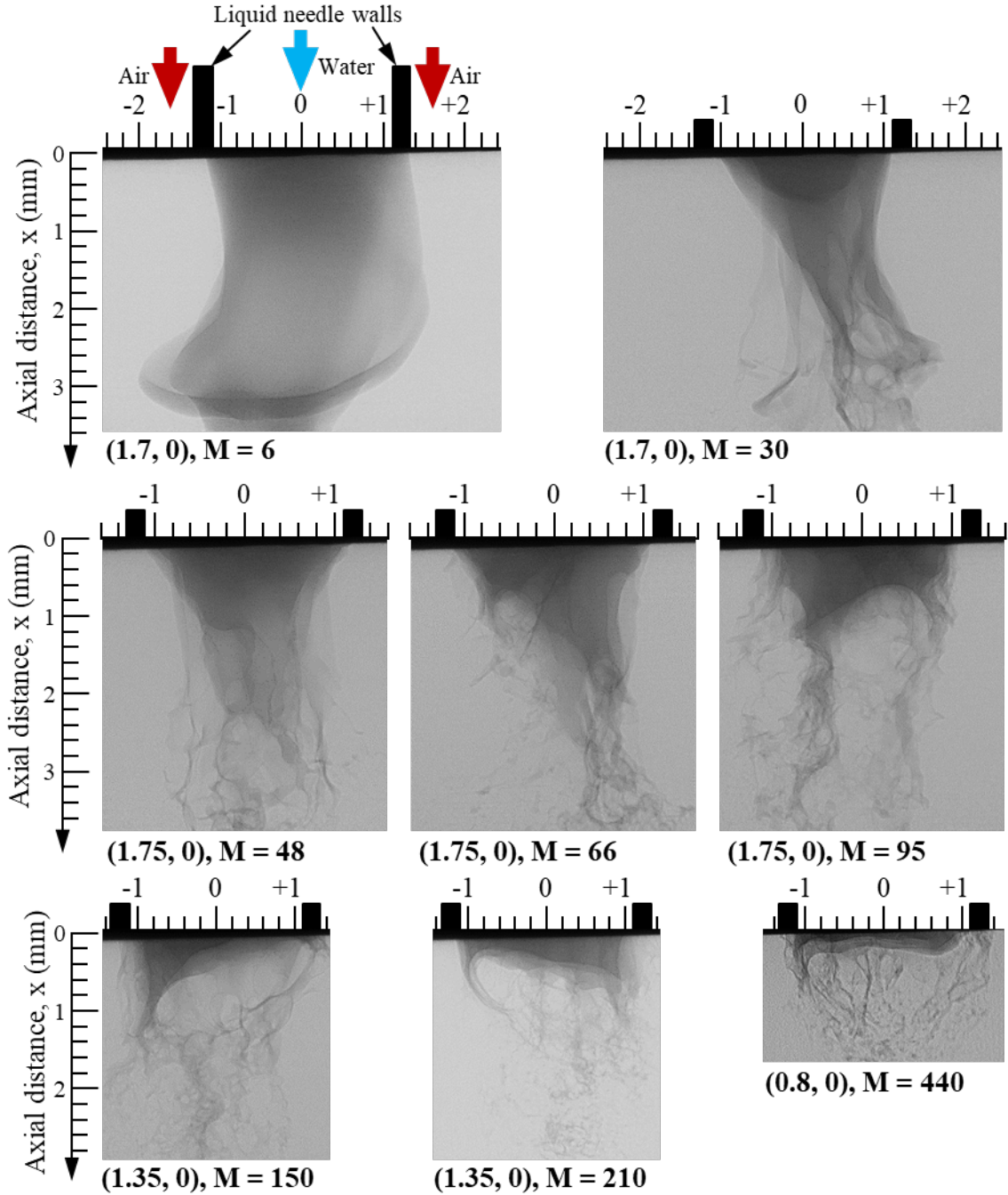


775

776 Figure 13: Secondary breakup at $M = 30$. Each image is separated by $278 \mu\text{s}$. Stretching of the

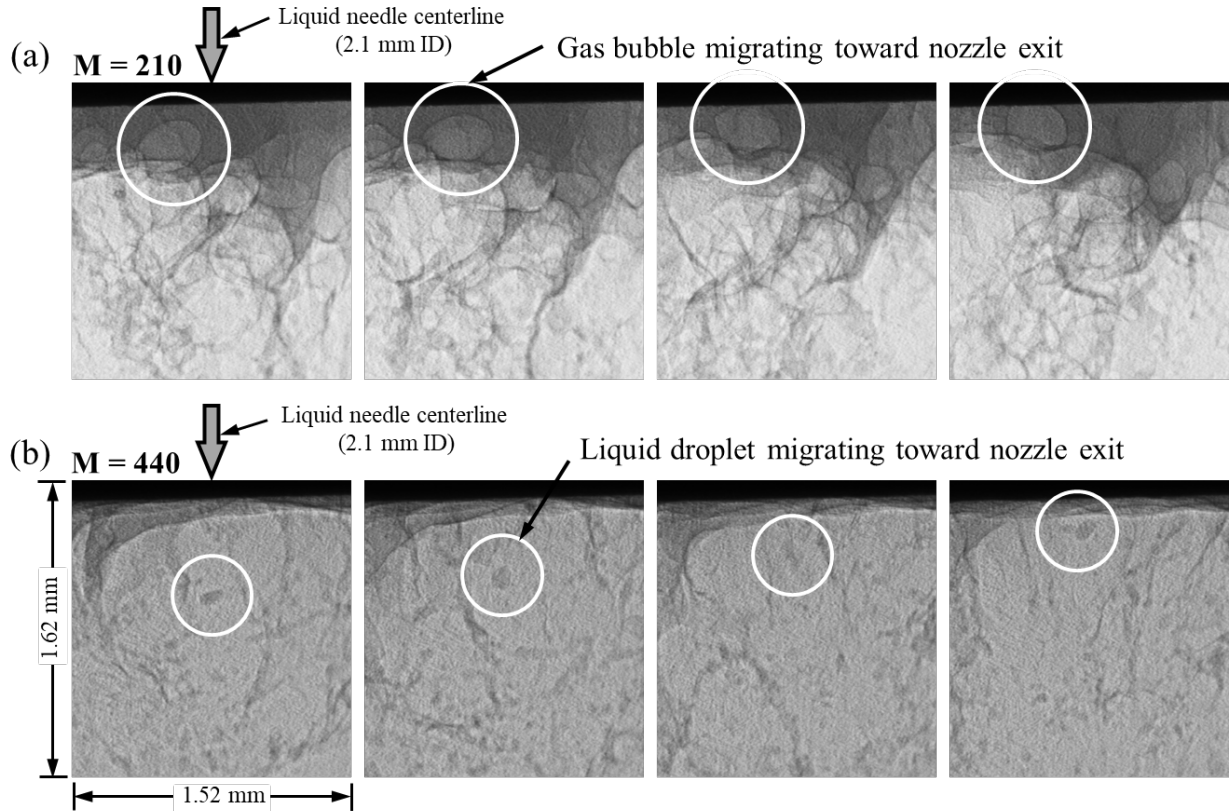
777 initial droplet into an oblong shape (A), which then undergoes bag formation (B) and

778 breakup (C), that leads to two resulting droplets (D).



779

780 Figure 14: White beam X-ray images right at the nozzle exit for various momentum flux ratios,
 781 while $SR = 0$ and the liquid flow rate is fixed at $Re_l = 1,100$. Specific flow conditions
 782 are summarized in Table 1. Coordinate locations correspond to the (x, y) image center
 783 and are given in mm, which also represents the scale in each image.



784

785 Figure 15: Close-up images taken at 100 kHz right at the exit of the liquid needle for

786 (a) $M = 210$ showing the upstream migration of an air bubble, and (b) $M = 440$

787 showing the upstream migration of a liquid droplet. The time between frames for each

788 sequence is $50 \mu\text{s}$.