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28 I. INTRODUCTION

29 Numerical and experimental investigations have demonstrated that the laser energy addition
30 technique is applicable to aircraft for improvement of their aerodynamic performances. Laser-induced
31 gas breakdown was first observed in 1963 [1-3]. Thereafter, the laser-induced gas breakdown was
32 believed to be applicable to a wide range of engineering applications such as flow control [4-6] and
33 laser plasma igniters [7, 8] to name a few. Flow control using laser-induced gas breakdown has been
34 shown to significantly contribute to drag reduction [9-11] and modification of shock waves [12-15].
35 Sperber *et al.* [16] demonstrated that a drag reduction of approximately 40% and 60% can be achieved
36 in Mach 2.1 and 2.7, respectively. Furukawa *et al.* [15] applied the laser energy deposition technique
37 to a supersonic free-flight model and suggested that shock wave modification due to laser energy
38 deposition has the potential to mitigate the sonic boom level. Tamba *et al.* [17] and Osuka *et al.* [18]
39 attempted to control shock-boundary layer interaction, which occurs on supersonic/hypersonic
40 vehicles, by repetitive-pulse laser energy deposition and showed that shock-boundary layer interaction
41 can be controlled.

42 Laser-induced gas breakdown is accompanied by a thermal spot and a blast wave. The thermal spot
43 and the blast wave play a key role in influencing the aerodynamic performances. A local high
44 temperature region corresponding to a low Mach number region induces shock wave movement when
45 the thermal spot interacts ahead of a shock wave. According to both numerical and experimental
46 investigations of laser energy addition ahead of a bow shock wave [11, 19, 20], the local high
47 temperature region modifies the shape of the bow shock wave. The local stand-off distance increases
48 in the vicinity of the thermal spot because of the relative low Mach number at the thermal spot.
49 Thereafter, a vortex ring is generated due to baroclinic effects and remains on a blunt body surface for
50 a period. The vortex ring is associated with a drag reduction [21, 22], and the residence time of the
51 vortex ring is proportional to the drag reduction [23]. Moreover, what is important is that the drag
52 reduction has a linear dependence on the thermal spot size as well as the ratio between the model
53 surface area interacting with the thermal spot and the thermal spot size. According to a numerical
54 investigation of the effect of laser energy deposition on a normal shock-boundary layer interaction in
55 a viscous duct flow [12], a blast wave induced by gas breakdown enhances the wall pressure behind
56 the separation point where the normal shock wave occurs. This pressure enhancement increases the
57 flow separation region, which is the length between the flow separation and reattachment points.
58 However, lower energy addition hardly influences the flow characteristics. The strength of the blast
59 wave and thermal spot size are the important parameter to provide sufficient effectiveness of laser
60 energy deposition.

61 The local thermal structure induced by the gas breakdown alters when the laser beam is focused in
62 any kind of impurities. The optical system for laser focusing greatly affects the laser discharge [24,
63 25]. Spherical aberration of the lens, the unfocused beam diameter, and the lens focal length all
64 influence the laser intensity profile along the beam path in the vicinity of the ideal focal spot [24]. The
65 breakdown threshold significantly depends on gas-medium, gas pressure, and the laser wavelength [26,
66 27]. In a pure gas, a long wavelength as well as a high medium pressure decrease the breakdown
67 threshold [28, 29]. The breakdown threshold in aerosols or any impurities shows the same tendency as
68 that in a pure gas; however, it is generally lower than the theoretical predictions and experimental
69 results [30-32]. Pinnick *et al.* [31] investigated effects of aerosols on breakdown thresholds at various
70 laser wavelengths and showed that breakdown thresholds are 2 to 3 orders of magnitude below those
71 for pure air. The breakdown threshold for a 10.6 μm laser beam continues to decrease with increasing
72 dust particle size at a range of approximately 0.1 to 100 μm [32, 33]. These results show that the lower
73 breakdown threshold is caused by laser-particle interaction. According to the review article from
74 Lushnikov *et al.* [34], the particle temperature is increased due to laser absorption, and thereafter a
75 higher temperature causes melting and evaporation of the particle. The particle shape may also change
76 due to laser absorption. Moreover, the laser energy absorption efficiency depends on the laser
77 wavelength, particle diameter, and particle temperature. Thus it is expected that the medium
78 gas/particle temperature distribution differs from a local thermal spot induced by gas breakdown in a
79 pure gas because of the higher number of parameters involved when particles are present. The different
80 thermal spot size and structure would affect the blast wave formation because rapid heating is
81 associated with blast waves.

82 Although previous investigations have shown that the laser-induced gas breakdown technique has
83 benefits for drag reduction and the control of shock wave structures, the importance of impurity effects
84 on the gas breakdown must be recognized before application to aircraft. This is because the laser beam
85 is not necessarily focused in a pure gas without impurities. Therefore, the effect of liquid impurities
86 on laser-induced gas breakdown is experimentally investigated in this study. Since the thermal spot as
87 well as the shock wave structure that is induced by gas breakdown are important factors for flow
88 control, the focus of this study is the investigation of the influences of impurities on the shock wave
89 structure as well as the low density region related to the thermal spot.

90 **II. EXPERIMENTAL SETUP**

91 A laser beam was focused in a quadrilateral stainless steel vacuum chamber with a volume of 0.138
92 m^3 . The chamber has a top quartz window of 98 mm in diameter, and a pair of side quartz windows
93 137 mm in diameter. The chamber was connected to a vacuum pump, and nitrogen gas was supplied

94 until the pressure within the chamber recorded 100 kPa after evacuating the air to $P_v = 20$ kPa. Based
95 on Dalton's law [35], the gas molecule consists of oxygen and nitrogen of approximately 4.2% and
96 95.8% in the chamber, respectively. Note that it is assumed that the atmospheric gas constituent is
97 oxygen at 21% and nitrogen at 79% in the chamber before evacuating the air. In the present
98 experiments, the pressure and the temperature in the chamber were $P_0 = 99.6 \pm 0.3$ kPa and $T_0 = 293.3$
99 ± 1.8 K, respectively. The nitrogen gas was passed through a particle generator (TSI, model: 9307-6,
100 1000 L/min of aerosol flow rate) to provide oleic acid based oil particles of 0.9 ± 0.63 μm in diameter
101 [36]. The supplied gas pressure was maintained at approximately 150 kPa. The volume of input
102 nitrogen gas is calculated using equation; $V_{in} = (M \cdot R \cdot T_0)/(P_0 - P_v)$, M and R are mass of nitrogen
103 gas and the gas constant, respectively. The mixture of nitrogen gas with suspended particles was
104 supplied into the chamber. According to an experimental result from Echols et al. [37], the Laskin
105 nozzle, which is installed into the particle generator we used, supplies particle mass concentration C_m
106 ≈ 4884 mg/m^3 in the present experimental condition. Therefore, particles with a total mass of $m_t =$
107 $C_m \cdot V_{in} = 150$ mg corresponding to 1×10^{-3} mg/cm^3 are suspended in the chamber. When considering
108 laser-induced gas breakdown without liquid impurity effects, the nitrogen gas was directly supplied to
109 the chamber. A pressure transducer (Kulite, model: XTE-190M, pressure range: 0 to 170 kPa Absolute)
110 driven by a DC power supply (TAim-TTi, model: EX752M) and k-type thermocouple were installed
111 on the chamber. These signals were monitored using a data acquisition system (National Instruments
112 Corp., model: NI-9205 for pressure monitoring and NI-9213 for temperature monitoring) driven by
113 LabVIEW.

114 To induce the laser energy focusing in the chamber, a Q-switched pulsed Nd: YAG laser beam with
115 a wavelength of $\lambda = 532$ nm was employed. The pulse width is $\tau_w \approx 10$ ns. A laser grade concave lens
116 with a focal length of -50 mm expands the laser beam and a second laser grade convex lens with a
117 focal length of 150 mm collimates the laser beam. The laser beam then passes through the top window
118 of the chamber. The unfocused collimated beam diameter was approximately $D = 25$ mm measured
119 using a photographic paper (ILFORD, model: MGIV Multigrade IV RC DELUXE, MGD.1M) before
120 the laser beam passes through the convex lens. The collimated laser beam is focused in the chamber
121 by a laser grade convex lens with a focal length of $f = 100$ mm which corresponds to a focusing f -
122 number $\equiv f / D = 4$. The laser energy in the vicinity of the laser focal spot was $E_{in} = 188$ mJ/pulse
123 measured using a power meter (Coherent Inc., model: PM USB LM-45).

124 The laser-induced blast wave and a low density core were visualised using high-speed schlieren
125 photography with a standard Z-type optical arrangement. The schlieren system consists of a continuous
126 light source with a 450 W Xe arc lamp (Newport, model: 66921), a condenser lens with a focal length
127 of 70 mm, an iris diaphragm, a pair of 203.3 mm diameter concave mirrors with a focal length of 1829

128 mm, a knife-edge, an imaging lens, and a high-speed camera either Fastcam SA1.1 (Photron, maximum
129 special resolution: 1024×1024 pixels) or HPV-1 (Shimadzu, special resolution: 312×260 pixels).
130 The iris in front of the condenser lens creates a light spot that illuminates the first parabolic mirror.
131 The light beam is then collimated by the first mirror and passes through the quartz side window of the
132 test section. A second parabolic mirror reflects the collimated beam after the beam passed through the
133 test section and another quartz side window. The knife-edge is horizontally positioned at the focal point
134 of the second parabolic mirror. The imaging lens in front of the camera focuses the image to the camera
135 sensor. The images were acquired at a frame rate of 37.5 kfps (Fastcam SA1.1) and 500 kfps (HPV-1)
136 with an exposure time of $1 \mu\text{s}$. An offset angle between the collimated light beam and the light path
137 from the light source to the first/second mirrors was set at 12 degrees to prevent coma aberration.

138 **III. RESULTS AND DISCUSSIONS**

139 **A. Shock wave structure**

140 Different shock wave formation/propagation become apparent in the laser-induced gas breakdown
141 with and without particles. Figure 1 shows the time evolution of schlieren images of the laser induced-
142 gas breakdown without liquid particles. The elapsed time after the breakdown is defined as t [μs]. The
143 laser beam is focused from the top in each image. Rapid local heating due to the laser focusing results
144 in plasma generation, which can be seen as the bright region in the schlieren images. A laser-induced
145 blast wave (LBW) propagates spherically into the surrounding gas. Schlieren images of the typical
146 shock wave structures in laser-induced gas breakdown with particles as well as the schematic of the
147 interaction are shown in Figs. 2 to 5. It seems that several shock waves are generated by multiple
148 breakdowns at the different breakdown locations along the beam path. The reason why multiple
149 breakdowns appear along the beam path will be discussed in the following paragraph. These shock
150 wave structures are categorized into four types; Category 1: the dual breakdowns (Fig. 2), Category 2:
151 the dual breakdowns with large energy consumption at a lower breakdown position (Fig. 3), Category
152 3: the dual breakdowns with large energy consumption at an upper breakdown position (Fig. 4),
153 Category 4: the triple breakdowns (Fig. 5),

154 In Category 1, dual breakdowns induced by similar laser energy consumption at two breakdown
155 points leads to two spherical shock waves. The shock wave fronts that propagate towards the opposite
156 breakdown point impinge on one another, and thereafter the transmitted shock waves (TSWs 1 and 2)
157 appear. Since the speed of sound in the heated spot is faster than that in the surrounding gas [38], the
158 TSWs travel through the heated spot faster, which results in a longitudinal elliptic shape in each blast
159 wave. In Category 2, the breakdowns are induced at two locations along the beam path but the laser

160 energy is relatively more consumed at the lower breakdown point. This results in a large shock radius
161 at the lower breakdown point. The shock wave structure in Category 3 is formed in an inverted position
162 of that in Category 2. In other words, the strong shock wave is generated at the upper breakdown point.
163 In Category 4 in which the triple breakdowns can be observed (Fig. 5), a middle breakdown generates
164 LBW 2 that interacts with LBW 1 and LBW 3, and thereafter the LBWs are coalesced at $\Delta t > 4 \mu\text{s}$.
165 The LBWs are transformed to the TSWs when the shock fronts imping on the others. Since the shock
166 radii of the LBWs 1 and 3 are smaller than that of LBW 2 at the elapsed time of $4 \mu\text{s}$, the coalesced
167 LBW forms a shuttle shape; however, the shuttle shaped LBW gradually transforms into a spherical
168 shape. It seems that the shock wave strength of the LBWs 1 and 3 is weaker than that of the LBW 2
169 because the high shock Mach number causes a larger shock radius for LBW 2 at the elapsed time of 4
170 μs .

171 The condition of suspended particles leads to the shock wave structures which were categorized
172 here into four types. In dual breakdowns (Category 1), the laser beam impinges on suspended particles
173 above the ideal focal spot, which results in breakdown due to laser-particle interaction. Even when the
174 laser energy is consumed at the location where the first breakdown occurred, the laser energy is still
175 sufficiently high so that another breakdown can be generated. The laser beam is focused towards the
176 ideal focal location thereafter, and the laser power density reaches the breakdown threshold due to the
177 narrow beam waist in the vicinity of the ideal laser focal spot. Even though the power density is lower
178 than the gas breakdown threshold in pure air, the impingement on the suspended particle causes
179 breakdown because the breakdown thresholds are 2 to 3 orders of magnitude below those for pure air
180 [40]. If the laser energy consumption at the upper breakdown location is the same as lower one, the
181 shock wave structure of Category 1 appears. The location of the particles interacting with the laser
182 beam alters the laser energy consumption at each breakdown point. In Category 2, the laser beam
183 impinges on fewer particles at the upper breakdown point, whereas more particles impinge on the
184 upper breakdown point in Category 3 compared with Category 2. Since particles are suspended
185 randomly, the occurrence of the shock wave structures of all Categories is a random event. Although
186 the shock wave structures observed here are categorized into four types, there is a possibility that
187 different shock wave structures can occur in different particle concentrations. Additionally, the laser
188 properties (wavelength and input energy etc.), the environment (gas pressure and gas molecule), and
189 particle properties (material and size) would cause different shock structures because they all influence
190 the breakdown threshold [26, 27, 30-34].

191 The breakdown appears at a lower laser energy density region due to laser-particle interaction.
192 Figure 6 shows the different laser focal locations for gas breakdown with and without particles. A
193 needle, the shadow on the right hand side in the image, is located in the vicinity of the ideal laser focal

194 spot so that the location of the multiple breakdown points can be detected. The ideal focal position
 195 corresponds to the centre of the LBW in gas breakdown without particles. In breakdown with particles,
 196 a longer longitudinal bright region related to plasma generation appears, which results in multiple
 197 laser-induced blast waves being produced in close proximity to one another (Figs. 2 to 5). Thereafter,
 198 a longitudinal elliptic shaped blast wave is formed due to the interaction of the multiple LBWs. As
 199 shown in Figs. 2 to 5, the multiple breakdown regions are only generated along the beam path. This
 200 allows us to expect that the laser beam with a narrow beam waist impinges on the particles around the
 201 ideal laser focal spot. In the breakdown with particles, the centre of the LBW is positioned at $z = 2.1$
 202 ± 0.2 mm above the ideal laser focal spot. The plasma can also generate at the upper part of the ideal
 203 focal spot. A possible scenario is that liquid particles melt due to local heating induced by the
 204 impingement of the laser beam with particles, and a vapour plume is formed due to vaporization from
 205 particle melting. Because the interaction between the vapour and the laser beam results in ionization
 206 of the gas [39], the laser-particle interaction induces a plasma. Based on the breakdown locations, the
 207 laser power density is estimated. The laser beam converges and diverges towards the ideal laser focal
 208 location. The breakdown without particles appears at the ideal focal spot, and the beam waist r_0 at the
 209 ideal focal spot is calculated as [40];

$$r_0 = r(0) = \frac{\lambda \cdot f}{\pi \cdot D/2} \quad (1)$$

210 It is assumed that the laser beam has a Gaussian intensity profile. In the case of suspended particles,
 211 the breakdown is generated above the ideal laser focal spot, and the location of the breakdown is
 212 sufficiently away from the ideal focal location ($z \gg z_R$). Thus the beam radius at which the breakdown
 213 appeared is expressed as [41];

$$r(z) \approx \frac{z \cdot \lambda}{\pi \cdot r_0} \quad (\text{for } z \gg z_R) \quad (2)$$

$$\text{where the Rayleigh range } z_R = \frac{f^2 \cdot \lambda}{\pi \cdot (D/2)^2}$$

214 Laser power density Q at the breakdown location is calculated using equation (3).

$$Q = \frac{E_{in}/\tau_w}{\pi \cdot r(z)^2} \quad (3)$$

215 Laser power densities with and without particle interaction cases are $Q \approx 5.5 \times 10^{11}$ and 3.3×10^{14}
 216 W/cm², respectively. The power density at which the breakdown appeared due to laser-particle
 217 interaction is significantly lower than that of without particle interaction. The laser-particle interaction
 218 induces breakdown even at the lower power density region.

219 **B. Shock radii and shock Mach number**

220 The shock radius and the shock Mach number were obtained from the schlieren images. The
221 longitudinal and lateral radii of the shock wave front are shown in Fig. 7. Figure 7 (b) shows the mean
222 value of all Categories of the shock wave structures. The longitudinal and lateral radii R_a and R_b
223 correspond to the opposite direction of the laser beam incidence and perpendicular to the beam path,
224 respectively. These radii are defined as the distance between the centre of the LBW and the outer shell
225 of the shock front (see sketch in Fig. 7). The centre of the LBW is measured from a range of the elapsed
226 time of 4 and 6 μs . This time is the first instant at which we can recognize the shock wave clearly. The
227 error bars of the shock radii in the breakdown with and without particles show the standard deviation
228 from 48 and 24 repetitions, respectively. The linear approximation curves of the shock radius are
229 estimated using the least-squares method. The Mach number of the shock wave fronts are calculated
230 from temporal variations of shock wave radii, assuming pure nitrogen gas in the chamber, the ambient
231 speed of sound is $C_0 = 349$ m/s. The shock Mach curves are obtained by logarithmic approximation.
232 The superscript “w” and “w/o” denote gas breakdown with and without particles, respectively.

233 Shock wave propagation in the lateral direction leads to the overpressure enhancement behind the
234 shock wave front due to multiple gas breakdowns. In gas breakdown without particles, all the shock
235 radii are almost the same. The longitudinal shock Mach number $M_a^{w/o}$ would be the same as the lateral
236 one $M_b^{w/o}$. Although the longitudinal and lateral shock Mach numbers are not completely the same in
237 the present results, the gradient of the approximation curves are similar. In gas breakdown with
238 particles, the longitudinal shock radius R_a^w is larger than R_b^w at the elapsed time of 4 μs because of
239 the elliptic shaped blast wave; however, the longitudinal and lateral shock radii become similar with
240 time. As shown in Figs. 2 to 5, the several shock waves induced by the longitudinal multiple
241 breakdowns constitute the elliptic shaped LBW. The lateral shock Mach number M_b^w is the fastest of
242 all the Mach numbers in both gas breakdown with and without particles. Since the shock Mach number
243 is proportional to the overpressure magnitude of the blast wave [42], the shock wave that propagates
244 towards the lateral direction leads to the large overpressure behind the shock front. As shown in the
245 sketches of Figs 2 to 5, the several shock waves interact with each other; however, the shock interaction
246 behaviour is different between the shock propagation in the longitudinal and the lateral directions. The
247 shock wave that propagates in the longitudinal direction passes through another shock wave, namely
248 the TSW. In contrast, the shock waves that propagate in the lateral direction would be
249 reflected/combined.

250 The triple breakdowns lead to a strong shock wave due to shock-shock interaction. Figure 8 shows
251 the comparison of the lateral shock Mach numbers in all the Categories. Approximation curves of

252 Categories 1 to 3 in which dual breakdowns occur are similar, whereas the triple breakdowns (Category
 253 4) generate a faster Mach number. In Category 4, three independent blast waves are induced by gas
 254 breakdown at early stage of the gas breakdown development, whereas two independent blast waves
 255 appear in Categories 1 to 3. Since the several blast waves interact with each other in Category 4, there
 256 is a higher possibility that more complicated shock interaction appears compared with the other
 257 Categories. The multiple shock interactions might cause the formation of a strong shock wave.

258 C. Laser energy efficiency

259 Laser-particle interaction leads to higher laser energy consumption. Based on the blast wave
 260 trajectory, the initial energy release can be theoretically predicted by a self-similarity solution well
 261 known as the Taylor-Sedov approach [43]; however, it is not applicable to weak blast waves. In other
 262 words, the Taylor-Sedov solution shows good agreement with only strong blast waves such as nuclear
 263 explosion [44]. The self-similarity solutions proposed by Sedov [45], Brode [46], and Jones [47],
 264 meanwhile, are applicable to the transition regime of an acoustic wave. Gebel *et al.* [48] applied the
 265 above three self-similarity solutions to detect the initial energy release of a weak blast wave and
 266 showed that both Brode's and Jones's methods successfully predict the initial energy release. Thus we
 267 apply the following Jones's method which is a semi-empirical approach [47] to estimate the initial
 268 amount of energy release;

$$\tau = a \left[\left(1 + b \cdot \varepsilon(t)^{\frac{n+2}{n}} \right)^{\frac{2}{n+2}} - 1 \right] \quad (4)$$

$$\varepsilon(t) = \frac{R_{pre}(t)}{R_0} \quad (5)$$

$$R_0 = \left[\left(\frac{n+2}{2} \right)^2 \cdot \frac{E_{bw}}{B \cdot \gamma \cdot P_0} \right]^{1/n} \quad (6)$$

$$t = \frac{(\tau \cdot R_0)}{C_0} \quad (7)$$

269 where τ is the non-dimensional time, a , b , and n are 0.543, 4.61, and 3 for a spherical blast wave
 270 respectively. $R_{pre}(t)$ and R_0 are shock radius from the theoretical prediction and a reference radius,
 271 respectively. The geometry parameter B , which is deduced by Jones [49], is 5.33 for the specific-heat
 272 ratio $\gamma = 1.4$. t is the arrival time at the position $R_{pre}(t)$. The blast wave energy E_{bw} that is consumed
 273 from the laser energy to generate the blast wave is estimated with an in-house program. The increment
 274 of the iterations is 0.1 mJ, and the minimum RMSE (Root Mean Squared Error) leads to the best fitting
 275 curve shown in Fig. 9. The RMSE is calculated as;

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (R_{exp}(t)_i - R_{pre}(t)_i)^2} \quad (8)$$

276 where $N = 13$ is the sampling number of the total measured experimental shock radius. R_{exp} is the
 277 experimental shock radius. It is assumed that the volume of the elliptic shaped blast wave corresponds
 278 to that of a circular blast wave, thus the experimental shock radius can be calculated as:
 279 $R_{exp}(t) \sim (R_b(t)^2 \cdot R_a(t))^{1/3}$. The RMSE of the fitting curves for $R_{exp}^{w/o}$ and R_{exp}^w are 0.126 and
 280 0.172, respectively. The best fitting curve allows to detect the blast wave energy E_{bw} . The laser energy
 281 efficiency $\eta = E_{bw}/E_{in}$ in gas breakdown with and without particles is shown in Table 1. The blast
 282 wave energy in breakdown with particles is 2.8 times higher than that without particles; hence, laser-
 283 particles interaction can improve laser energy efficiency in creating a blast wave. Note that the present
 284 experimental results show that E_{bw} in gas breakdown without particles has a relatively lower laser
 285 energy efficiency compared with previous investigations [48, 50] because an optical compartments
 286 and the parameters of the laser beam such as input laser energy, laser absorption coefficient, and the
 287 focusing f -number etc. all influence the laser energy efficiency [40, 50].

288 **D. Low density distribution**

289 The local thermal spot leads to a special low density form called a kernel. Figures 10 and 11 show
 290 the sequential images of typical kernel formation with and without particles. These images are captured
 291 using the Photron camera. Figure 11 is categorized as the dual breakdowns (Category 1). In gas
 292 breakdown without particles (Fig. 10), the kernel is a lateral elliptic shape at the elapsed time of $27 \mu s$
 293 although the kernel is almost the circular in shape at $\Delta t < 27 \mu s$ because the schlieren image captured
 294 using Shimadzu camera shows the circular shaped kernel at the time of $10 \mu s$ in Fig. 1 (d). Thereafter,
 295 the bottom region of the kernel moves towards the opposite direction of the laser beam incidence
 296 because of the asymmetric temperature distribution [51].

297 The local low density spot becomes larger in breakdown with the presence of particles. The extreme
 298 high gas temperature due to the laser focusing causes the particles surrounding the laser focal spot to
 299 melt, which results in the wide low density field around the laser focal location. This is because the
 300 high temperature particle, due to laser energy absorption, heats the surrounding particles. In breakdown
 301 with particles (Fig. 11), a longitudinal elliptic shaped kernel is formed because the two independent
 302 plasma regions generated along the beam path in Category 1. In a single laser focusing [51-53], a
 303 vortex ring, which circulates towards the opposite direction of the laser incidence, is generated due to
 304 a pressure gradient. The dual breakdowns in which two vortex rings appear leads to these vortices

305 interaction (Fig. 11 (b) and (c)). As shown in Fig. 11, the turbulent structure can be observed, which
306 results in gas mixing enhancement. Note that the kernel formation depends on both the breakdown
307 location and the number, thus the low density distribution in the other Categories will be discussed in
308 the next paragraph. According to the experimental results of Glumac *et al.* [54], the local gas
309 temperature rises up to approximately 50,000 K at the laser focal spot of the 532 nm laser beam with
310 150 mJ/pulse. Thus we can expect that a similar thermal spot may appear in present experiments
311 although the laser energy, laser intensity distribution, and unfocused collimated beam diameter are not
312 the same as Glumac's experiment. Considering temperature in Glumac's experiments, it can deduce
313 that the high temperature has the capability to widely melt the particles surrounding the laser focal
314 spot. As previously discussed in the fourth paragraph, Subsection III A (Shock wave structure), the
315 location of the breakdown moves slightly up from the ideal focal position, thereby the laser beam with
316 a larger beam waist has a much higher probability to interact with the vapour plume caused by particle
317 melting. In other words, a wider plasma region appears compared with gas breakdown without
318 particles. This results in the low density spot having much wider distribution.

319 The volume of the thermal spot and the number are a key factor for the kernel formation in multiple
320 breakdowns. It is deduced that the shape of the thermal spot would be altered depending on Category
321 of the shock structure. This is because multiple breakdowns appear in suspended particles, thereby
322 there is a higher possibility that the thermal spots interact with each other. The various kernel
323 formations in breakdown with particles are shown in Fig. 12. The dual breakdowns (Category 1) lead
324 to two low density cores with a similar volume along the beam path. Since these low density cores
325 occur close to each other, the gas mixing is enhanced at the interaction region. In the dual breakdowns
326 with large energy consumption at the lower breakdown (Category 2), the two low density cores are
327 formed along the beam path; however, the upper density core is smaller than the lower one. This is
328 because less laser energy is deposited at the upper breakdown compared with that at the lower one,
329 thereby the higher temperature spot is smaller. The upper low density core deforms relatively faster
330 than the lower core. This smaller density core is affected by the shock interaction. The transmitted
331 shock wave (TSW2, see Fig. 3), which propagates from the lower breakdown position, longitudinally
332 extends the upper kernel region due to a Richtmyer-Meshkov like instability. In the dual breakdowns
333 with large energy consumption at an upper breakdown (Category 3), the large kernel is formed at the
334 upper breakdown because of the higher laser energy deposited at this location. Due to the interaction
335 of the transmitted shock wave (TSW1, see Fig. 4), the lower small kernel is extended towards the
336 direction of the beam incidence. The small kernel development due to the shock interaction was not
337 necessarily be observed in both Categories 2 and 3. Even when the shock structure is categorized as
338 either Category 2 or 3, the present results show that the small kernel extension is hardly observed. The

339 reason for this is that the small kernel might be combined with the large kernel when the dual
340 breakdowns occur and the location of the breakdowns is too close. In the triple breakdowns (Category
341 4), three low density cores are formed and interact with each other. The lower and upper density cores
342 are extended due to the Richtmyer-Meshkov like instability. The transmitted shock waves (TSWs 1
343 and 3, see Fig. 5), meanwhile, impinge at both upper and lower parts of the middle density core, this
344 in turn causes the collapsed shape of the middle density core.

345 The vortex motion and instability at the upper/lower kernel region in the multiple breakdowns is
346 due to a pressure gradient induced by the shock interaction. According to an experimental investigation
347 of laser energy deposition at a single focal point [55], the plasma quickly expands and propagates
348 towards the opposite direction of the beam incidence at an early stage of the plasma development, i.e.,
349 the high gas temperature spot related to the plasma moves towards the direction of the laser beam
350 source. This results in the teardrop-shaped temperature distribution with a large volume as well as a
351 higher temperature at the focal lens side. The initial temperature distribution generates the blast wave
352 which propagates outward, and a local rapid temperature rise causes a strong blast wave. This results
353 in an expansion wave that produces a pressure gradient towards the centre of the blast wave [56]
354 because the outward shock motion induces an overexpansion of the inner gas [51]. In the multiple
355 breakdowns (Category 2) for example, the temperature distribution induces a vortex ring at the upper
356 kernel region, and thereafter the TSW2 interacts with the kernel corresponding to lighter fluid than the
357 surrounding gas. Since shock propagation is faster in the heated spot (kernel), the faster velocity behind
358 the TSW2 would induce the moving of the surrounding gas into the upper kernel (arrows in Fig. 3).
359 This enhances vortex motion and leads to instability. According to Hawley et al. [57], the inclined
360 interface between the lighter and heavy gases induces the instability of a vortex layer, hence the
361 instability would appear in present experiment because the contact surface of different density is
362 complicated.

363 Breakdown with particles in which multiple low density cores appear leads to a large volume of the
364 kernel. Figure 13 shows time history of the kernel formation, x and y are defined as the longitudinal
365 and lateral lengths shown in Fig. 10 (d), respectively. These lengths were measured from the schlieren
366 images captured using the Shimadzu camera. The error bars of the lengths in the breakdown with and
367 without particles show the standard deviation from 48 and 24 repetitions, respectively. Both $x^{w/o}$ and
368 x^w gradually increase with time but x^w is longer than $x^{w/o}$ because the wide plasma region generates in
369 the breakdown with particles. Both $y^{w/o}$ and y^w have the same tendency of the longitudinal length
370 decreasing then increasing slightly. It is expected that the time range between 40 and 50 μ s is a
371 transition region where the growth direction of the longitudinal length changes. In gas breakdown
372 without particles, both $x^{w/o}$ and $y^{w/o}$ have a narrow standard deviation, whereas a larger standard

373 deviation appears in gas breakdown with particles. In particular, the longitudinal length y^w has a higher
 374 standard deviation because both the laser focal location and the number of breakdowns alter greatly
 375 along the longitudinal direction; hence, they strongly influence the longitudinal length.

376 Based on the longitudinal and lateral radii of the kernel $r_x = x/2$ and $r_y = y/2$, the volume rate of the
 377 kernel $V_{rate} = V_{pk}^w/V_{pk}^{w/o}$ is calculated (Fig. 14). It is assumed that the kernel has an elliptic shape at
 378 the elapsed time between 12 and 45 μ s, thus its volume can be calculated as: $V_{pk} = 4/3 \cdot \pi \cdot r_x^2 \cdot r_y$.
 379 The error bars of volume rate σ_{rate} are estimated using the following equation;

$$\sigma_{rate} = \sqrt{\left(\frac{\partial f}{\partial r_x^{w/o}} \cdot \sigma_x^{w/o}\right)^2 + \left(\frac{\partial f}{\partial r_y^{w/o}} \cdot \sigma_y^{w/o}\right)^2 + \left(\frac{\partial f}{\partial r_x^w} \cdot \sigma_x^w\right)^2 + \left(\frac{\partial f}{\partial r_y^w} \cdot \sigma_y^w\right)^2} \quad (9)$$

380 where the volume rate is a function of $r_x^{w/o}$, $r_y^{w/o}$, r_x^w , and r_y^w . The subscripts “x” and “y” denote the
 381 lateral and longitudinal directions, respectively. The standard deviation σ is obtained based on the radii
 382 of the kernel. The mean value shows that the low density region in the breakdown with particles is in
 383 the range of 4 and 5.5 times as large as that without particles.

384 IV. CONCLUSION

385 The impurity effect of suspended liquid particles on laser-induced gas breakdown was
 386 experimentally investigated in quiescent gas. Laser-induced gas breakdown was generated in an
 387 environmental chamber filled with nitrogen gas with suspended oleic acid based oil particles $0.9 \pm$
 388 0.63μ m in diameter. To induce the laser energy deposition, a Q-switched pulsed 532 nm Nd: YAG
 389 laser beam with an output energy of 188 mJ/pulse was focused in the chamber.

390 The schlieren images showed that different shock wave formation/propagation are present in laser-
 391 induced gas breakdown with and without particles. Several shock waves were generated by multiple
 392 breakdowns along the laser beam path. A possible scenario is that the liquid particles melt due to the
 393 local heating induced by the impingement of the laser beam on the particles, thereby the laser-particle
 394 interaction induces a plasma even at the low power density region.

395 The shock wave structures are categorized into four types; Category 1: dual breakdowns, Category
 396 2: dual breakdowns with large energy consumption at the lower breakdown, Category 3: dual
 397 breakdowns with large energy consumption at the upper breakdown, Category 4: triple breakdowns.

398 The triple breakdowns in which three independent blast shock waves interact with each other
 399 enhanced the shock wave strength behind the shock front in the lateral direction. The shock wave fronts
 400 that propagate towards the opposite breakdown position impinged on one another, and thereafter a

401 transmitted shock wave (TSW) appeared. The TSW interacted with the low density core called the
402 kernel. The kernel then expanded due to a Richtmyer-Meshkov like instability; however, the kernel
403 formation depended on the breakdown location and the number of breakdowns. Laser-particle
404 interaction caused the increase in the kernel volume, and the volume of the kernel in the breakdown
405 with particles was approximately five times as large as that in the gas breakdown without particles. In
406 addition, laser-particles interaction can improve laser energy efficiency in generating a blast wave
407 since the blast energy was 2.8 times higher than in gas breakdown without particles.

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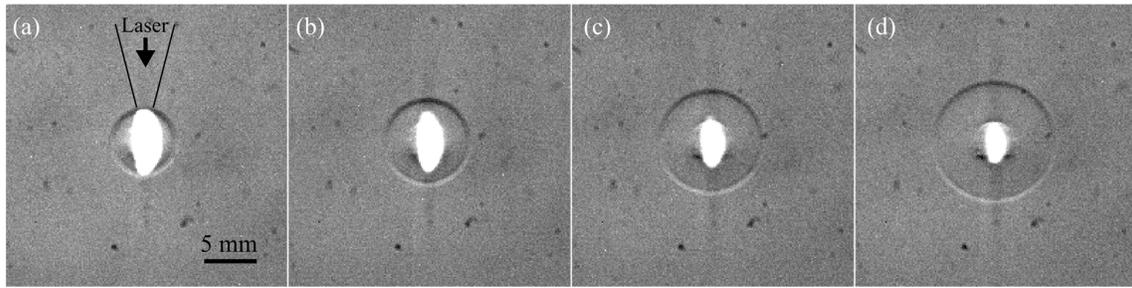
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534 **Figures**

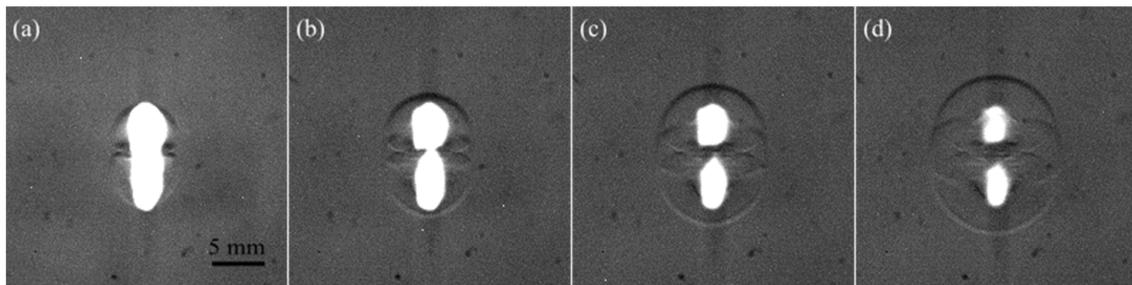


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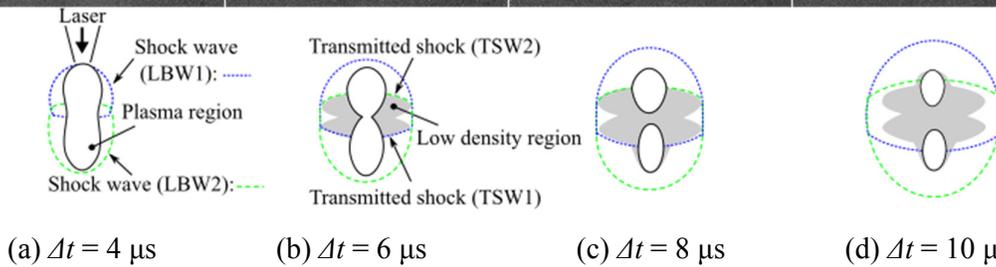
(a) $\Delta t = 4 \mu s$ (b) $\Delta t = 6 \mu s$ (c) $\Delta t = 8 \mu s$ (d) $\Delta t = 10 \mu s$
 Figure 1. Typical sequential schlieren images of laser-induced gas breakdown without particles



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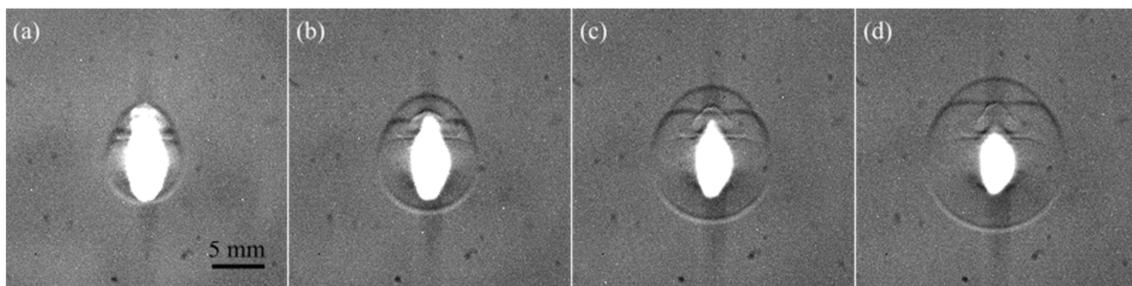
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(a) $\Delta t = 4 \mu s$ (b) $\Delta t = 6 \mu s$ (c) $\Delta t = 8 \mu s$ (d) $\Delta t = 10 \mu s$

Figure 2. Laser-induced gas breakdown with particles, Category 1: the dual breakdowns

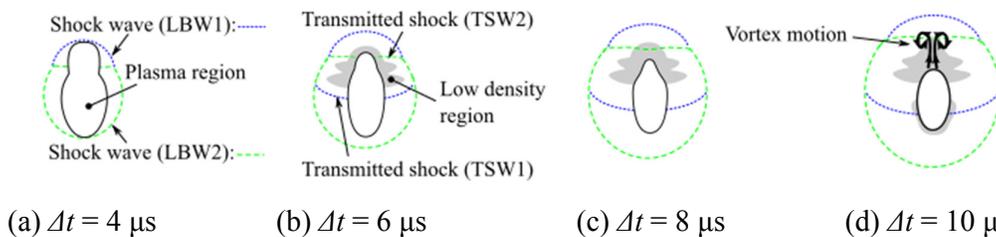


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(a) $\Delta t = 4 \mu s$ (b) $\Delta t = 6 \mu s$ (c) $\Delta t = 8 \mu s$ (d) $\Delta t = 10 \mu s$

Figure 3. Laser-induced gas breakdown with particles, Category 2: the dual breakdowns with large energy consumption at the lower breakdown position

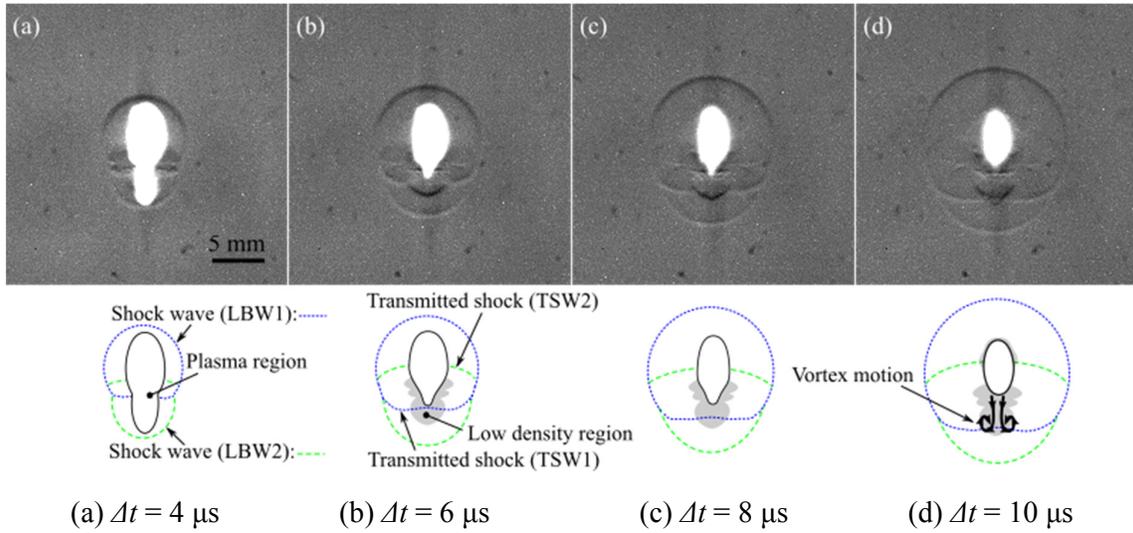


Figure 4. Laser-induced gas breakdown with particles, Category 3: the dual breakdowns with large energy consumption at the upper breakdown position

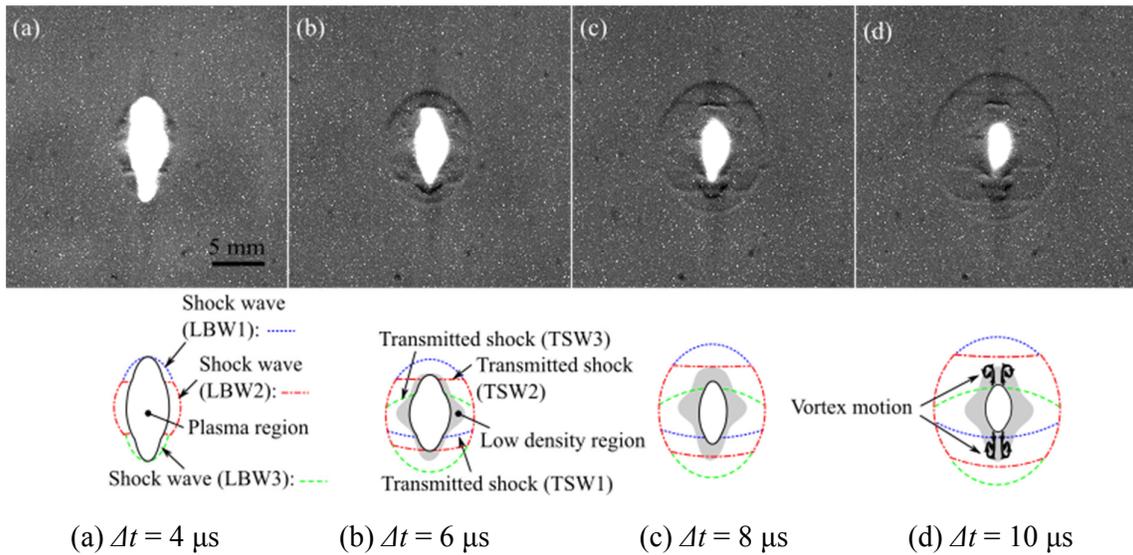


Figure 5. Laser-induced gas breakdown with particles, Category 4: the triple breakdowns

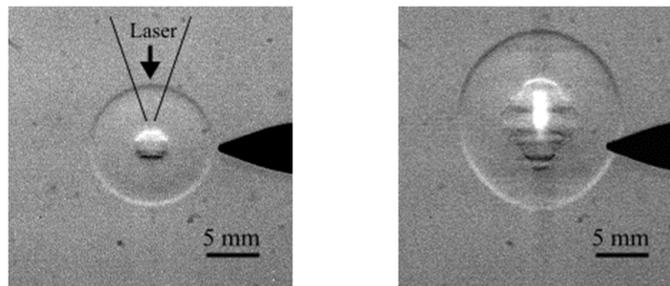
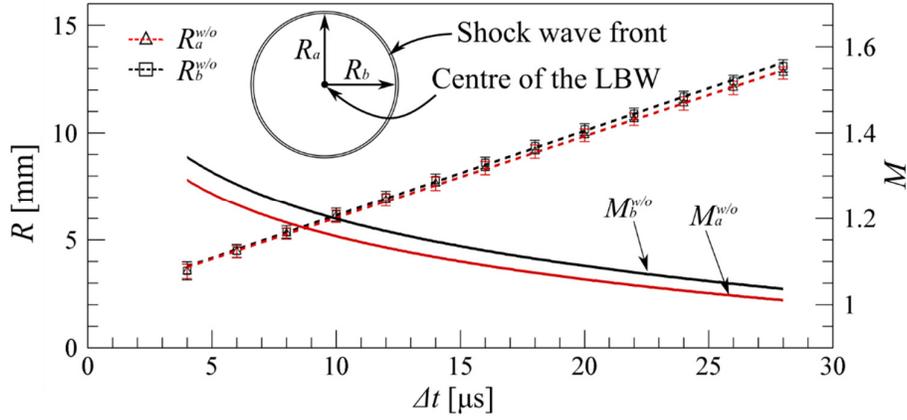


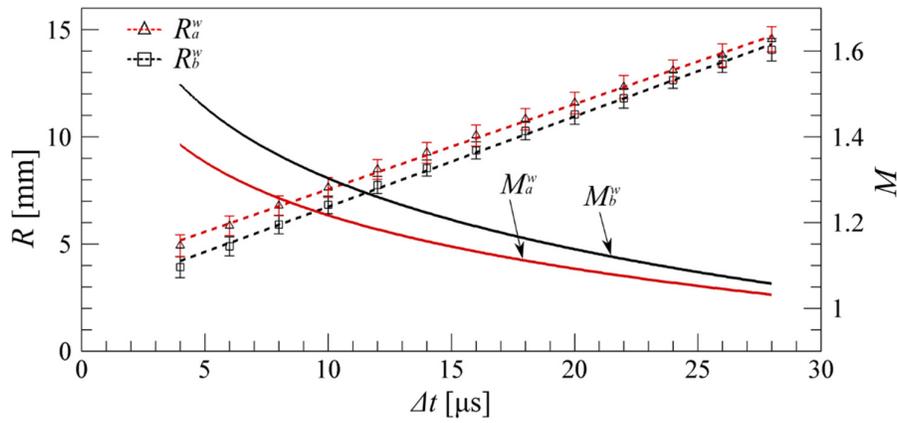
Figure 6. Typical laser-induced gas breakdown at the elapsed time of $10 \mu\text{s}$



555

556

(a) without particles



557

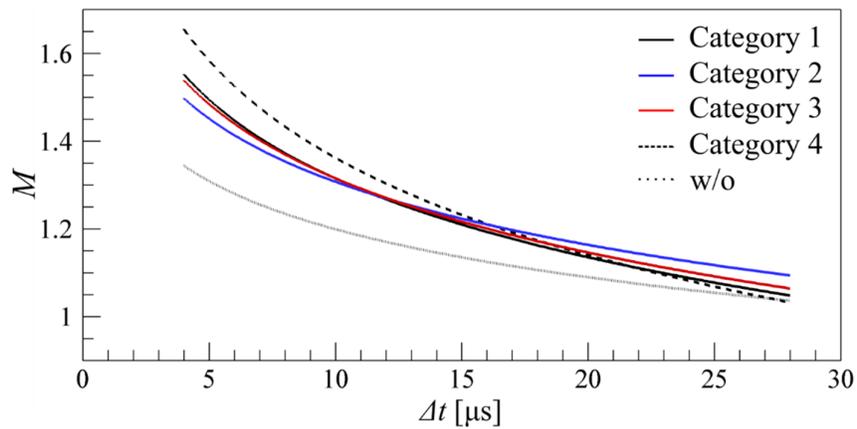
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(b) with particles

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Figure 7. The longitudinal and lateral radii of the laser-induced blast wave and Mach number of the shock wave front



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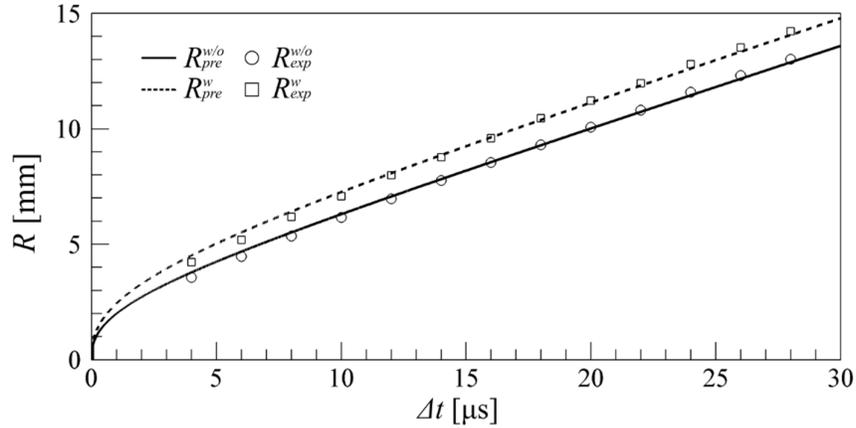
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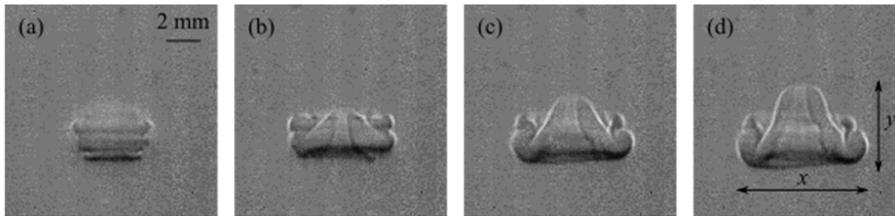
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Figure 8. The Mach number of the lateral shock wave front at the various categories, Category 1: the dual breakdowns, Category 2: the dual breakdowns with large energy consumption at a lower breakdown position, Category 3: the dual breakdowns with large energy consumption at an upper breakdown position, Category 4: the triple breakdowns



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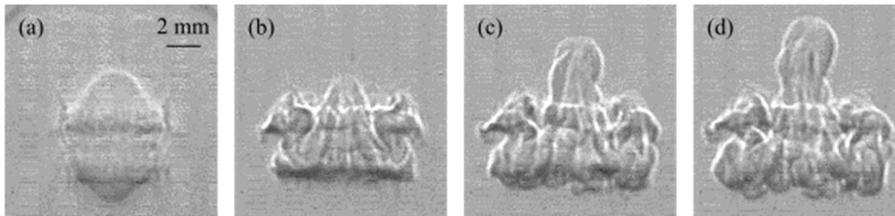
567 Figure 9. Comparison of the experimental shock radius with the theoretical fitted curve, solid and
 568 dashed lines denote the best fitting curve by theoretical predilection method



569

570 (a) $\Delta t = 27 \mu s$ (b) $\Delta t = 108 \mu s$ (c) $\Delta t = 189 \mu s$ (d) $\Delta t = 270 \mu s$.

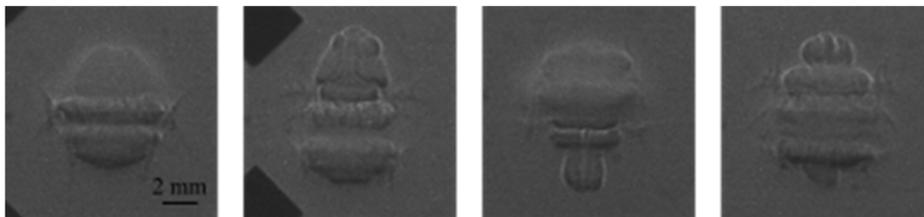
571 Figure 10. Temporal variation of the kernel formation in gas breakdown without particles



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573 (a) $\Delta t = 27 \mu s$ (b) $\Delta t = 108 \mu s$ (c) $\Delta t = 189 \mu s$ (d) $\Delta t = 270 \mu s$

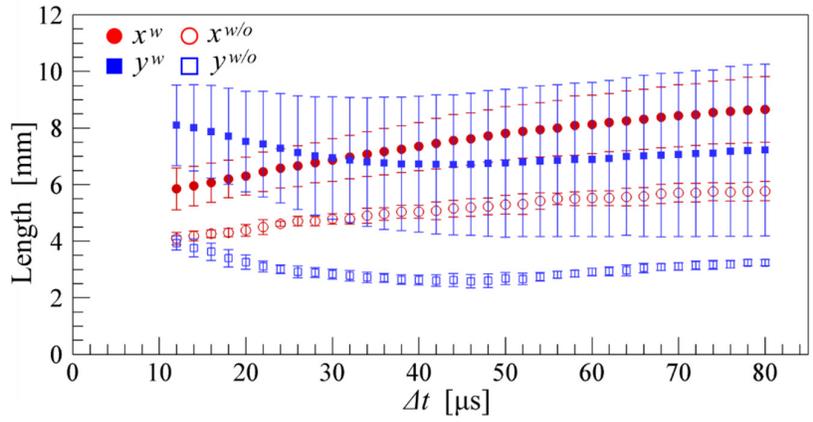
574 Figure 11. Temporal variation of the kernel formation in gas breakdown with particles (Category 1)



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576 (a) Category 1 (b) Category 2 (c) Category 3 (d) Category 4

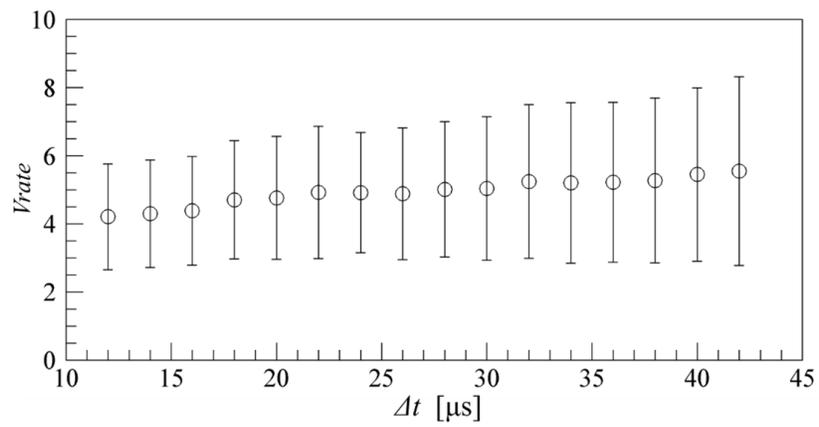
577 Figure 12. Various kernel formations at the elapsed time of 18 μs , Category 1: the dual breakdowns,
 578 Category 2: the dual breakdowns with large energy consumption at a lower breakdown position,
 579 Category 3: the dual breakdowns with large energy consumption at an upper breakdown position,
 580 Category 4: the triple breakdowns



581

582

Figure 13. Temporary variation of the longitudinal and lateral lengths of the kernel



583

584

Figure 14. Volume rate of the low density spot

585 **Table**

586 Table 1. Laser energy efficiency in breakdown without and with particles

	Without particles	With particles
$\eta = E_{bw}/E_{in}$ [%]	14	39

587