

Melt flow instability in laser metal additive manufacturing

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14 This work is supported by US National Science Foundation. This research used resources of the
15 Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility
16 operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-
17 AC02-06CH11357.

18 CRediT author statement

19 **Qilin Guo:** Conceptualization, Formal analysis, Investigation, Methodology, Writing – original
20 draft, Writing – review & editing. **Minglei Qu:** Investigation, Methodology, Validation. **Luis I.**
21 **Escano:** Investigation. **S. Mohammad H. Hojjatzadeh:** Investigation. **Zachary Young:**
22 Investigation. **Kamel Fezzaa:** Methodology, Resources. **Lianyi Chen:** Conceptualization,
23 Funding acquisition, Investigation, Methodology, Project administration, Resources, Writing –
24 review & editing.

25 Declaration of interest

26 The authors declare no competing interests.

27 **Revealing melt flow instabilities in laser powder bed fusion additive manufacturing of**
28 **aluminum alloy via in-situ high-speed X-ray imaging**

29

30 **Abstract**

31 Laser metal additive manufacturing technologies enable the fabrication of geometrically and
32 compositionally complex parts unachievable by conventional manufacturing methods. However,
33 the certification and qualification of additively manufactured parts are greatly hindered by the
34 stochastic melt flow instabilities intrinsic to the process, which has not been explicitly revealed by
35 direct observation. Here, we report the mechanisms of the melt flow instabilities in laser powder
36 bed fusion additive manufacturing process revealed by in-situ high-speed high-resolution
37 synchrotron X-ray imaging. We identified powder/droplet impact, significant keyhole oscillation,
38 and melting-mode switching as three major mechanisms for causing melt flow instabilities. We
39 demonstrated the detrimental consequences of these instabilities brought to the process, and
40 projected new understanding on the melt flow evolution and keyhole oscillation. This work
41 provides critical insights into process instabilities during laser metal additive manufacturing,
42 which may guide the development of instability mitigation approaches. The results reported here
43 are also important for the development and validation of high-fidelity computational models.

44 **Keywords**

45 Additive manufacturing, laser processing, melt flow, synchrotron X-ray imaging

46

47 **1. Introduction**

48 Laser metal additive manufacturing technologies have the potential to revolutionize
49 manufacturing industry by enabling the fabrication of geometrically and compositionally complex
50 parts unachievable by conventional manufacturing methods [1–3]. To fabricate parts with
51 desirable and predictable quality, extensive research have dedicated to correlate the process
52 dynamics (melt pool variation [4,5], pore formation [6–8], spatter generation [9,10], keyhole
53 oscillation [11–13], etc.) with the processing conditions (laser power, scan speed, beam size, etc.),
54 in an effort to establish an “optimized” set of parameters to produce parts with less defects and

55 higher density [14–17]. However, there are uncertainties intrinsic to the laser metal additive
56 manufacturing process where some unstable physical dynamics are not tightly bonded to specific
57 processing conditions [16–20]. Such instabilities pose great uncertainty to the qualification and
58 certification of the additively manufactured parts [20–22], which require explicit characterization
59 through direct observations.

60 To investigate the process instabilities, it is essential to trace the transient melt flow behavior
61 inside the melt pool—the direct product of laser-matter interaction. However, the opacity of metals
62 to visible light poses great barrier in direct observation of the molten metals within the melt pools.
63 To overcome this challenge, recent research has applied synchrotron radiation based in-situ X-ray
64 imaging to observe the physical dynamics within metals [5,6,10,23–26]. By in-situ X-ray imaging,
65 the localized melt flow behavior within a laser induced metallic melt pool could be inferred from
66 the movement of pores generated during the process [5,26,27]. The regular melt flow patterns
67 within the whole melt pool have also been studied using tungsten particles as flow tracers [6,23–
68 25].

69 Limited research has been conducted toward experimental investigations on melt flow
70 instabilities. In blown powder directed energy deposition (DED) additive manufacturing process,
71 it was reported that the impact of feeding particles can cause melt pool surface fluctuations,
72 generate porosity, and cause keyhole oscillations [26]. In laser powder bed fusion (LPBF) additive
73 manufacturing process, it was reported that high laser scan speeds and large powder layer thickness
74 can cause unstable melt flow, which could lead to rough surface finish [28]. The melt flow behavior
75 was inferred by the morphology of solidified track, as well as the powder spattering behavior.
76 Recent research using in-situ X-ray imaging to monitor the LPBF process has reported several
77 defect-formation mechanisms resulting from unstable melt flow behavior or depression zone
78 fluctuations, although the unstable melt flow behavior itself was not characterized [29].

79 Computational modeling work has also been performed to study the melt flow instabilities. In
80 general, the studies focused on two aspects: the instability formation mechanism and the
81 consequences of the instabilities on the process. Surface tension variation was identified as a
82 source of melt flow instabilities, as surface tension is one of the major driving forces for liquid
83 migration. The surface tension fluctuations could be induced by both improper processing
84 parameters (such as hatch spacing [30]) and chemical composition variations (such as increased

85 oxidation level [31]) . The inhomogeneous powder packing in the LPBF powder bed also serves
86 as a source to disturb the melt flow by cutting off the liquid migration at the loose-packing region,
87 resulting in part defects such as porosity and balling [32]. As for the consequences, the melt flow
88 instabilities have been reported to be accountable for the breakup of melt tracks (Plateau-Rayleigh
89 instability), trap of gas pores, and creating denudation zone around the keyhole rim during LPBF
90 [33]. Other melt flow induced process instabilities such as liquid ejection and periodical
91 oscillations of keyhole have also been demonstrated by high-fidelity simulations [11,34,35].

92 So far, in-process experimental characterization of the melt flow instabilities during LPBF has
93 not been reported. In the present work, we report the melt flow instabilities within aluminum melt
94 pools during laser powder bed fusion process revealed by in-situ high-speed, high-energy, high-
95 resolution synchrotron X-ray imaging with uniformly dispersed populous micro-tracers. We
96 investigate the mechanisms for causing three major types of melt flow instabilities and quantify
97 the influence of these instabilities in both local scale and global scale. We also demonstrate the
98 detrimental consequences that the instabilities exert to the process. Inspired by the results, we
99 further elaborate our new understandings on the mechanisms of keyhole oscillation and melt flow
100 evolution.

101 **2. Methods and Materials**

102 **2.1 In-situ laser melting X-ray imaging experiment**

103 We used in-situ laser-melting X-ray imaging to monitor the dynamics of melt flow inside the
104 melt pool during laser melting/scanning on an aluminum powder bed, as schematically illustrated
105 in Fig. 1(a). The powder bed was composed of a metal substrate (0.5 mm thick along X-ray
106 transmission direction), a manually-spread powder layer with 100 μm thickness, and two glassy
107 carbon walls for holding the powder. A vertical Gaussian laser beam with a $1/e^2$ diameter of ~ 100
108 μm scans the powder bed to create a moving melt pool. The laser is a 1070 nm wavelength,
109 continuous-wave, single-mode, ytterbium fiber laser (YLR-500-AC, IPG Photonics, USA),
110 positioned by a galvo scan head (IntelliSCAN_{de} 30, SCANLAB GmbH, Germany). During laser
111 scanning, a stationary high-energy synchrotron X-ray beam (at beamline 32-ID of Argonne
112 National Laboratory's Advanced Photon Source) penetrated through the specimen from horizontal
113 direction. The transmitted X-ray beam carrying melt flow information was converted by a
114 scintillator (LuAG:Ce) into visible light, which was recorded by a high-speed camera with a frame

115 rate of either 140 kHz or 50 kHz, and a spatial resolution of 1.97 μm per pixel. Therefore, all the
116 physical dynamics were projected on a 2D imaging plane. Aluminum alloy feedstock powder
117 (AlSi10Mg and Al6061) were uniformly mixed with 1 vol.% flow tracers (5 μm tungsten particles)
118 by ball milling to trace the melt flow, as schematically illustrated in Fig. 1(b). The powder size
119 distribution of aluminum feedstocks after ball milling is shown in Fig. 1(c).

120 **2.2 Materials**

121 Two aluminum alloys were used in this work: AlSi10Mg and Al-6061. Aluminum alloys were
122 chosen for their high X-ray transparency. AlSi10Mg alloy, as one of the most widely used alloy in
123 additive manufacturing, has enhanced laser absorption by the enriched Si content [36]. Therefore,
124 AlSi10Mg was used in this work to study the melt flow in relatively large melt pools (keyhole-
125 mode and transition-mode). Al-6061, as a common aluminum alloy on market, has low laser
126 absorptivity. It was used to investigate the dynamics in conduction-mode melt pool or the incidents
127 that are sensitive to laser absorption.

128 The alloy substrates with dimensions of 40 mm \times 3 mm \times 0.5 mm for in-situ X-ray imaging were
129 prepared by wire electrical discharge machining (wire EDM). The dimension along X-ray
130 incidence is 0.5 mm to ensure better X-ray transparency. The surface of the substrate was ground
131 by 400-grit sand paper to remove any contaminations. The aluminum powders were uniformly
132 mixed with 1 vol.% tungsten micro-particles (5 μm) as flow tracers by ball milling.

133 **2.3 Surface morphology characterization**

134 Scanning electron microscopy (SEM) was performed on a Zeiss LEO-1530 field emission
135 scanning electron microscope to observe the solidified track surface morphology. The sample was
136 pre-tilted to 60° with respect to the electron beam for better observation of the track height.

137 The surface profiles of the solidified track were measured on a VHX-5000 Digital Microscope
138 (KEYENCE Corporation of America).

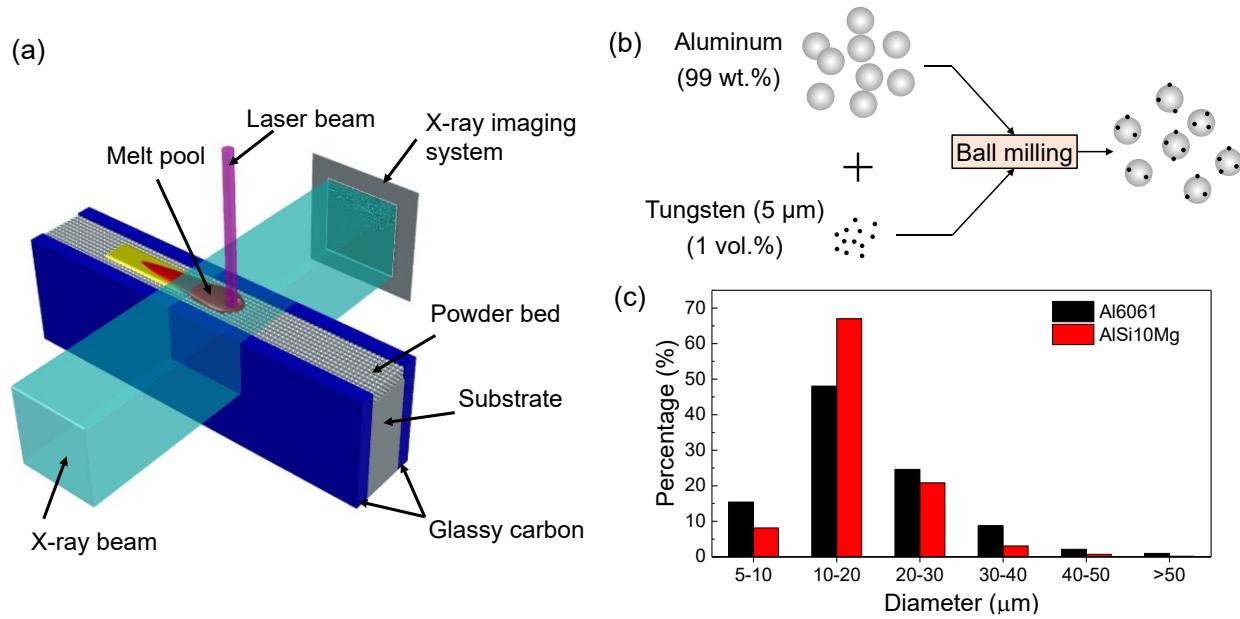
139 **2.4 Melt flow tracing approach**

140 The speed (v) of a tungsten tracer was calculated by dividing its displacement (d) by its traveling
141 time (t), $v = d/t$. The tracer's displacement (d) was calculated via its two-dimensional (2D)
142 coordinates change ($\Delta x = |x_2 - x_1|$, $\Delta y = |y_2 - y_1|$) from one frame to the next frame in the 2D X-ray

143 image planes, where $d = (\Delta x^2 + \Delta y^2)^{1/2}$. The tungsten tracer's travelling time (t) is the time interval
 144 between two frames, determined by the recording frame rate of the X-ray imaging video (50 kHz
 145 or 140 kHz in the present work).

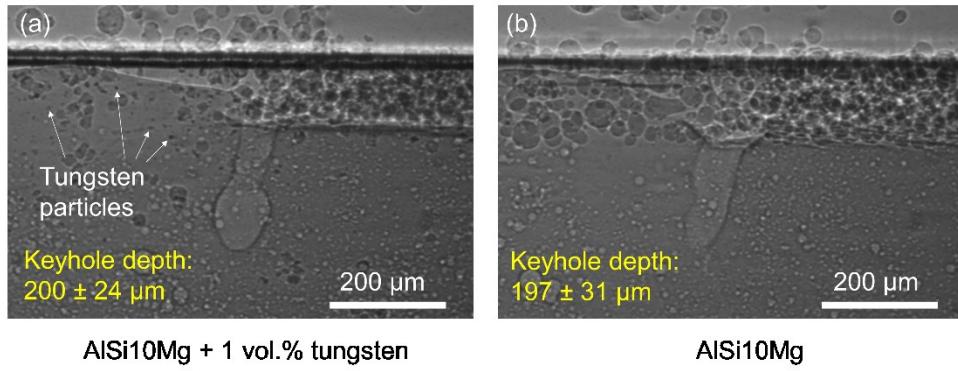
146 To examine whether adding 1 vol.% tungsten particles to aluminum powder bed could change
 147 the laser absorption behavior, we conducted two laser-melting experiments using different powder
 148 beds but identical laser processing conditions (364 W, 0.5 m/s). The results are displayed in Fig.
 149 2. As shown in Fig. 2(a), AlSi10Mg powder mixed with 1 vol.% tungsten particles generated a
 150 keyhole (laser induced vapor cavity) depth of $200 \pm 24 \mu\text{m}$ (averaged over 100 frames). The
 151 keyhole generated with pure AlSi10Mg powder bed exhibited an average depth of $197 \pm 31 \mu\text{m}$,
 152 as shown in Fig. 2(b), which is only 1.5% smaller than the keyhole depth generated with mixed
 153 powder bed. Therefore, the influence of 1 vol.% tungsten on laser absorption is minimal.

154 In addition to the laser absorption, it has also been reported that adding 1 vol.% of tungsten
 155 particles to the aluminum feedstock does not have significant impact on the physical property of
 156 the aluminum melt pool [23]. Thus, it is feasible to use tungsten microparticles as flow tracers.



157

158 **Fig. 1. Method for in-situ melt flow mapping experiment.** (a) Schematic illustration of the
 159 experiment setup for X-ray imaging of laser powder bed fusion process. (b) Powder preparation
 160 method for melt flow tracing. The feedstock aluminum powder was mixed with 1 vol.% tungsten
 161 particles by ball milling. (c) Aluminum particle size distributions of the feedstock Al6061 and
 162 AlSi10Mg powder after ball milling with tungsten particles. The distribution calculation did not
 163 include aluminum particles smaller than 5 μm or tungsten particles.



164

165 **Fig. 2. Comparison of keyhole depth during laser melting of AlSi10Mg + 1 vol. % tungsten mixed**
 166 **powder and pure AlSi10Mg powder. The laser processing conditions are identical for the two**
 167 **experiments: 364 W laser power, 0.5 m/s scan speed.**

168 **2.5 Definition of laser melting modes**

169 The laser power and scan speed were varied to realize three major melting modes during the
 170 investigation of melt flow instabilities, including keyhole mode, conduction mode, and transition
 171 mode. There are two major approaches to distinguish different melting modes.

172 One of the approaches is based on physics [37,38]: Keyhole-mode melting is dominated by
 173 convective heat transfer, conduction-mode melting is dominated by heat conduction; while
 174 transition-mode melting is in between of the keyhole mode and conduction mode.

175 The other classification approach is based on geometry [4]: A keyhole-mode melt pool contains
 176 large melt volume with a deep depression zone induced by intensive vaporization of materials. The
 177 aspect ratio $(W/2)/D$ (half width over depth) of the depression zone is usually less than 1. A
 178 conduction-mode melt pool forms under low laser radiation, thus contains small melt volume
 179 without having a depression zone. A transition-mode melt pool is created under conditions
 180 between keyhole-mode and conduction-mode laser melting, with a slightly larger (or similar) melt
 181 volume than conduction-mode melt pool, yet still holds a depression zone with an aspect ratio of
 182 $(W/2)/D > 1$. In the present work, we took the second approach to define the laser melting modes.

183 **3. Results**

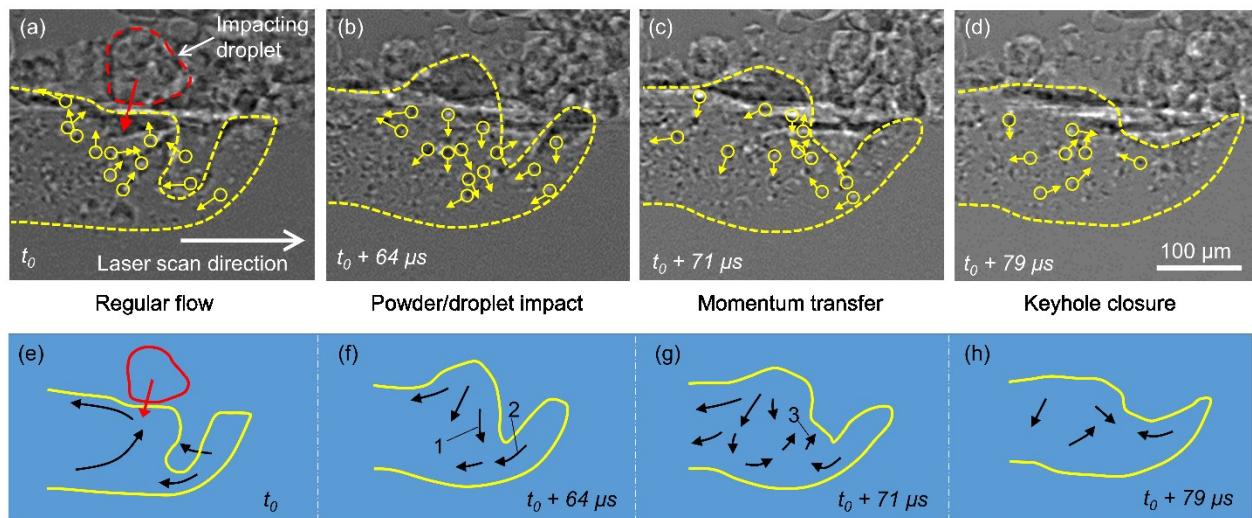
184 We identified three major mechanisms for causing melt flow instabilities during laser processing,
 185 i.e., powder/droplet impact, significant keyhole oscillation, and melting-mode switching. The
 186 influences of these instabilities on the melt flow behavior are demonstrated below.

187 **3.1 Powder/droplet impact induced melt flow instability**

188 The flowable powder, as the core and unique element in the dominating powder-based laser
 189 metal additive manufacturing technologies, enables great flexibility for process design, but also
 190 brings frequent disturbances to the process [13]. Herein, we report two types of melt flow
 191 instabilities induced by the powder.

192 **3.1.1 Local instability induced by powder/droplet impact**

193 In laser metal additive manufacturing process, a laser-driven proceeding melt pool continuously
 194 captures the powder on the powder bed to grow into a part. However, the incorporating powder
 195 can be large in size (more than three times larger than the feedstock powder), due to
 196 agglomerations or merging of small droplets. The impact of large powder clusters or droplets into
 197 a melt pool with large melt volume (i.e. keyhole-mode melt pool) could locally disturb the regular
 198 melt flow pattern, as elucidated in [Fig. 3](#) (and Supplementary Video 1).



199 **Fig. 3. Powder/droplet impact induced local melt flow instability.** (a-d) X-ray images showing
 200 the melt flow change during a droplet impacting to keyhole-mode melt pool. The laser power is
 201 312 W with a scan speed of 0.6 m/s. The material is AlSi10Mg. (e-h) Schematic illustration of the
 202 melt flow change in (a-d).

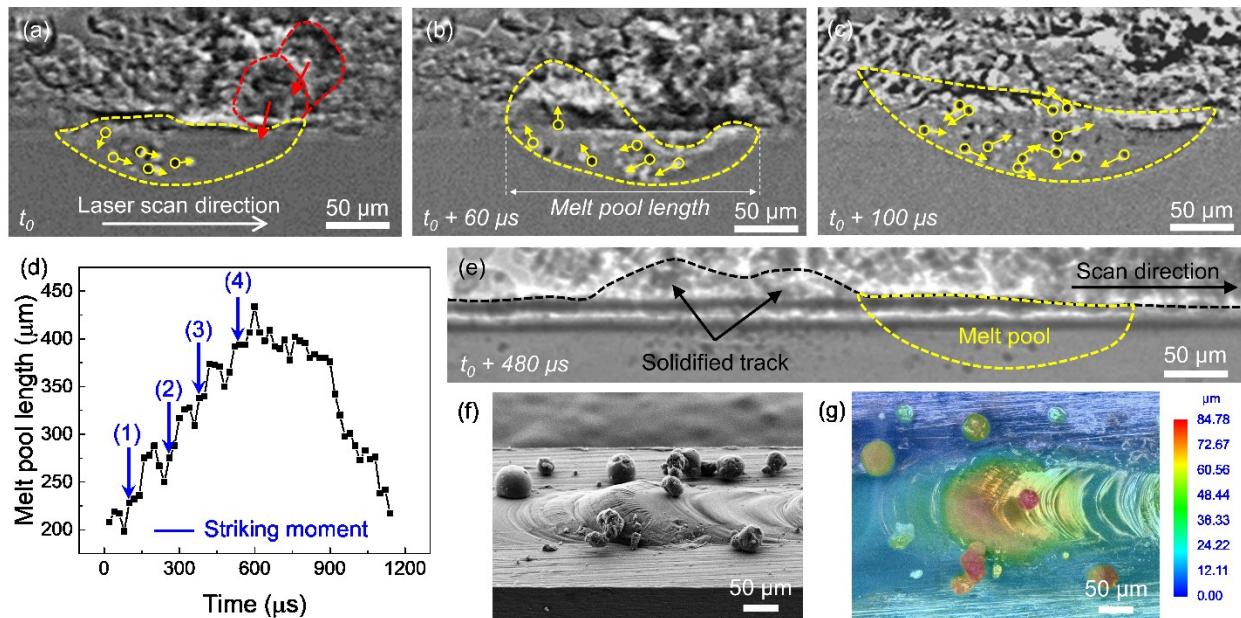
204 [Figure 3\(a-d\)](#) display X-ray images where a melt pool moves from left to right in the field of
 205 view. Yellow dashed lines marked the melt pool boundaries. The laser is invisible in the view,
 206 whereas its location was indicated by the moving keyhole. The flow tracers were circled with

207 arrows pointing out their instant moving directions. By connecting the movements of individual
 208 tracers, the melt flow patterns were deducted and schematically exhibited in Fig. 3(e-h).

209 During an impact, the droplet transfers kinetic energy and potential energy into the melt pool,
 210 locally altering the original flow direction (Fig. 3(a,e)) into the droplet momentum direction (Fig.
 211 3(b,f)) at the impact location. The collision between foreign flow (carrying liquid from the droplet)
 212 and the original flow (carrying liquid from the melt pool) exhausted the impact energy and
 213 dampened the droplet impact from spreading further. The downward flows 1 and 2 (Fig. 3(f))
 214 collided at the keyhole bottom and formed an upward flow 3 (Fig. 3(g)), pushing the keyhole
 215 bottom surface upward till the depression almost vanished (Fig. 3(d,h)). Although keyhole
 216 vanishing is momentary, it can reduce the local laser absorption and cause undesired energy
 217 fluctuation in the process [4].

218 3.1.2 Global instability induced by powder/droplet impact

219 Compared with the above keyhole-mode laser melt pool, a conduction-mode laser melt pool
 220 contains much less liquid volume, which cannot efficiently dampen and confine the
 221 powder/droplet impact within a local region. Rather, the melt flow instability brought by the
 222 impact on conduction-mode melt pool is global and more detrimental, as demonstrated in Fig. 4.



224 **Fig. 4. Powder/droplet impact induced global melt flow instability.** (a-c) X-ray images showing
 225 the melt flow change during a droplet impacting to conduction-mode melt pool. The laser power

226 is 312 W with a scan speed of 0.6 m/s. The material is Al6061. (d) Effect of droplet striking on the
227 melt pool length development as a function of time. (e) X-ray image showing the profile of the
228 solidified melt track resulting from droplet impact. (f) SEM image showing the solidified track at
229 the same region as in (e). (g) Surface profile of the solidified track at the same imaging area as in
230 (e) and (f).

231 During an impact, as shown in [Fig. 4\(a\)](#), two droplets together carrying a liquid volume nearly
232 one-third of a conduction-mode melt pool struck on the front melt pool surface. The impact broke
233 the original regular flow pattern in the whole melt pool, as evidenced by the reversed flow direction
234 at the rear-bottom of melt pool, which changed from moving forward ([Fig. 4\(a\)](#)) to backward ([Fig.](#)
235 [4\(b\)](#)). The surface level at the rear melt pool was kicked up by the striking ([Fig. 4\(b\)](#)) and rapidly
236 solidified as it is ([Fig. 4\(e,f\)](#)), adding up to the surface roughness of the as-printed layer. Surface
237 profiling measurement in [Fig. 4\(g\)](#) shows the elevated track height can be 50 μm -higher than the
238 average solidified track height. The impact droplet also increases the volume of liquid metal in the
239 melt pool, leading to the melt pool elongation, as shown in [Fig. 4\(c\)](#).

240 Large powder/droplet impact is not an occasional event in laser metal additive manufacturing
241 process. We quantified the striking incidence by evaluating the melt pool length change in 1200
242 μs during laser scanning, as shown in [Fig. 4\(d\)](#). Within the first 600 μs , we observed four striking
243 events, leading to a continuous elongation of the melt pool from $\sim 200 \mu\text{m}$ to 434 μm (over 100%
244 increase). We noticed that the droplet-striking event did not elongate the melt pool immediately.
245 The elongation usually occurs 20-60 μs after the striking, because the striking liquid takes time to
246 travel along the melt pool. No striking event happened for the remaining 600 μs (from 600 μs to
247 1200 μs in [Fig. 4\(d\)](#)), during which the melt pool length gradually recovered to the original size.
248 This result demonstrates that the powder/droplet striking occurs frequently and randomly during
249 laser scanning, which brings uncertainty to the qualification of additively manufactured parts.

250 **3.2 Significant keyhole oscillation induced melt flow instability**

251 3.2.1 Local instability induced by significant keyhole oscillation

252 The melt flow patterns around the keyhole are highly dependent on the keyhole behavior. A
253 significant keyhole oscillation with an amplitude over twice as large as the original keyhole size
254 can override the original flow patterns at adjacent areas.

When a significant keyhole oscillation happens, the liquid at the rear keyhole wall was pushed backward to form a surface wave, as shown by the X-ray images in [Fig. 5\(a,b\)](#) and the schematic illustrations in [Fig. 5\(e,f\)](#). The wave front I ([Fig. 5\(f\)](#)) squeezed the rear rim of the keyhole to generate a protruding surface wave, which propagated backward against the laser scanning direction. Aside from the main surface wave (as shown in [Fig. 5\(b\)](#)), a secondary wave possibly locating at the side of the melt track formed afterward, as displayed in [Fig. 5\(c\)](#) and [Supplementary Fig. 1](#). The possible configuration that could cause overlaying contrast in X-ray images is demonstrated in [Supplementary Fig. 2](#). The liquid beneath the surface wave got compressed and spread away to a broader area with a speed of ~ 0.6 m/s (measured by tracing the displacement of wave front II in [Fig. 5\(g,h\)](#)). With the spreading of compressed wave, the liquid metal at the affected area moved along the wave propagating direction temporarily, while the original flow pattern was temporarily erased and overridden.

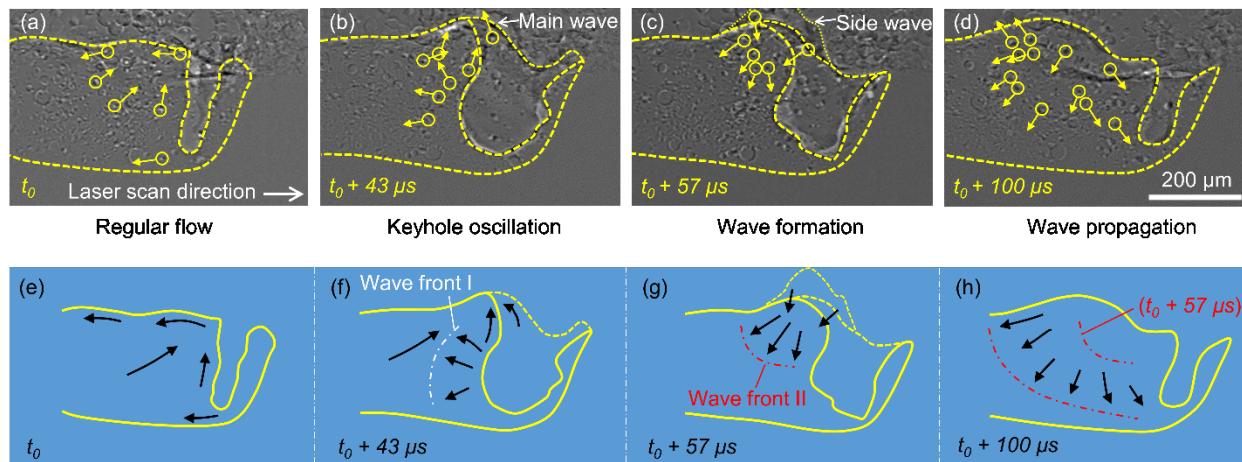


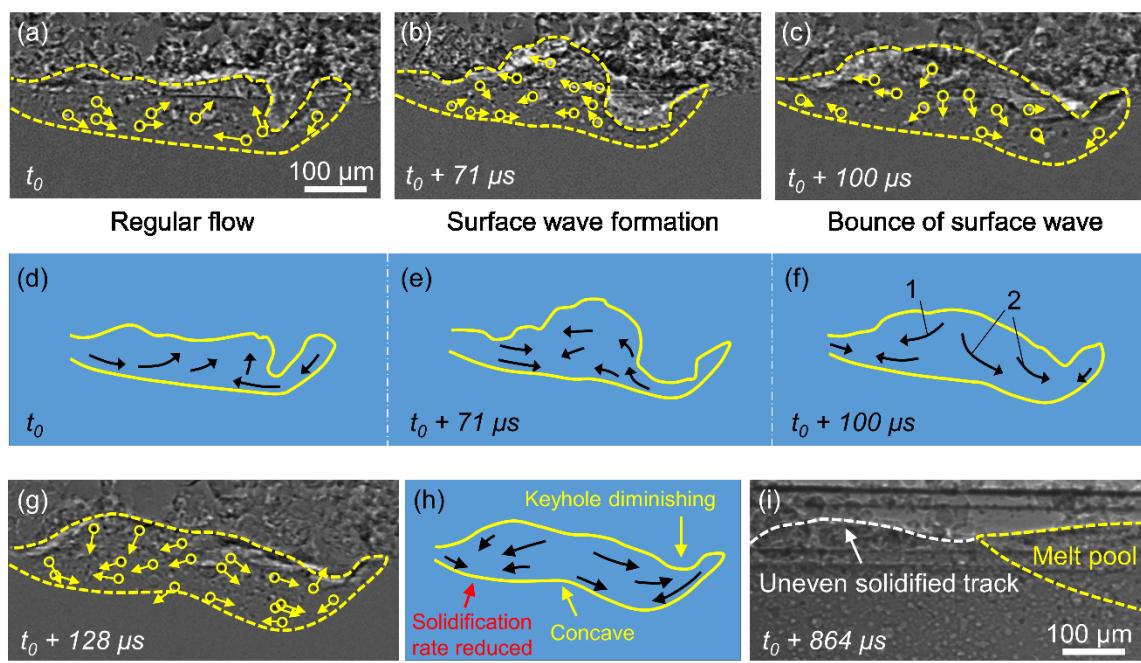
Fig. 5. Significant keyhole oscillation induced local melt flow instability. (a-d) X-ray images showing the melt flow change during a significant keyhole oscillation event. The laser power is 364 W with a scan speed of 0.6 m/s. The material is AlSi10Mg. (e-h) Schematic illustration of the melt flow change in (a-d).

3.2.2 Global instability induced by significant keyhole oscillation

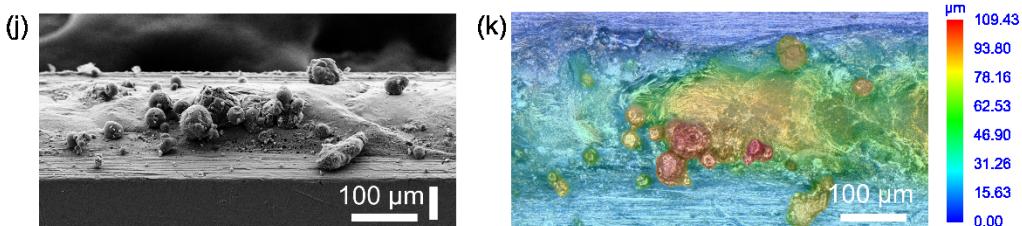
A global effect takes place when a significant keyhole oscillation occurs in a moderate-size keyhole-mode melt pool or a transition-mode melt pool, as shown by [Fig. 6](#). Initially, the oscillation created a backward-moving wave that compressed the liquid behind the keyhole, as shown in [Fig. 6\(b,e\)](#), which is similar to the event in [Fig. 5](#). However, different from [Fig. 5](#), the compressed liquid did not spread far before it touched the bottom of the shallow melt pool, where

278 the liquid split into two flows (Fig. 6(c,f)): flow-1, moving backward to the rear melt pool; and
 279 flow-2, moving forward to the front melt pool. The splitting flows initiated a series of instabilities
 280 to the process, as demonstrated in Fig. 6(g-i):

281 (1) Reduction of solidification rate occurred at the tail of the melt pool, as indicated by the red
 282 arrow in Fig. 6h. Under regular melt flow patterns, as shown in Fig. 6(a)), the solid-liquid interface
 283 at the melt pool bottom was smooth and convex. However, the significant keyhole oscillation
 284 pushed a large volume of liquid moving backward to the rear melt pool, slowing down the
 285 solidification at the rear bottom of the melt pool. As a result, a concave was observed on the solid-
 286 liquid interface at the middle of the melt pool bottom, as pointed out in Fig. 6(h). The original X-
 287 ray image of Fig. 6(b) without labelling the melt pool boundary was provided in Supplementary
 288 Fig. 3 to illustrate the visibility of melt pool solid-liquid interface.



289 (1) Reduced solidification rate; (2) keyhole closure; (3) melt pool elongation; (4) uneven surface



290 **Fig. 6. Significant keyhole oscillation induced global melt flow instability.** The laser power is
 291 312 W with a scan speed of 0.6 m/s. The material is AlSi10Mg. (a-c) X-ray images showing the
 292 formation and propagation of an abnormal surface wave. (d-f) Schematic illustration of the melt

293 flow pattern in (a–c). (g, i) X-ray images showing the consequences induced by the melt flow
294 instability. (h) Schematic illustration of the melt flow pattern in (g). (j) SEM image of the solidified
295 track in (i). (k) Surface profile of the solidified track in (i) and (j).

296 (2) The keyhole cavity was filled up by the forward-moving flow, as shown in Fig. 6(g,h),
297 following a similar mechanism as the flow-colliding-induced keyhole closure revealed in Fig. 3(f–
298 h).

299 (3) The solidification at the tail of the melt pool was delayed by the backward-moving flow, due
300 to the extra mass of molten alloy transported to the rear area. As a result, the melt pool elongated
301 from $605 \pm 9 \mu\text{m}$ to $691 \pm 10 \mu\text{m}$ during the event.

302 (4) An uneven surface of the solidified track was left when the surface wave reached the tail of
303 the melt pool and solidified with an elevated liquid level, as exhibited in Fig. 6(i,j). The surface
304 roughness is characterized by surface profiling, as shown in Fig. 6(k). The highest point at the
305 uneven solidified track is $\sim 40 \mu\text{m}$ higher than the average height of the solidified track.

306 So far, we have revealed the melt flow instabilities induced by powder/droplet impact and
307 significant keyhole oscillation. Their individual effects on various size of melt pools are
308 demonstrated to be different. In brief, the instabilities occurring in a large melt pool tend to
309 influence a portion of the melt pool, while those occurring in a relatively small melt pool usually
310 trigger a global reaction to the whole melt pool and can be more detrimental to the process.

311 **3.3 Melting-mode switching induced melt flow instability**

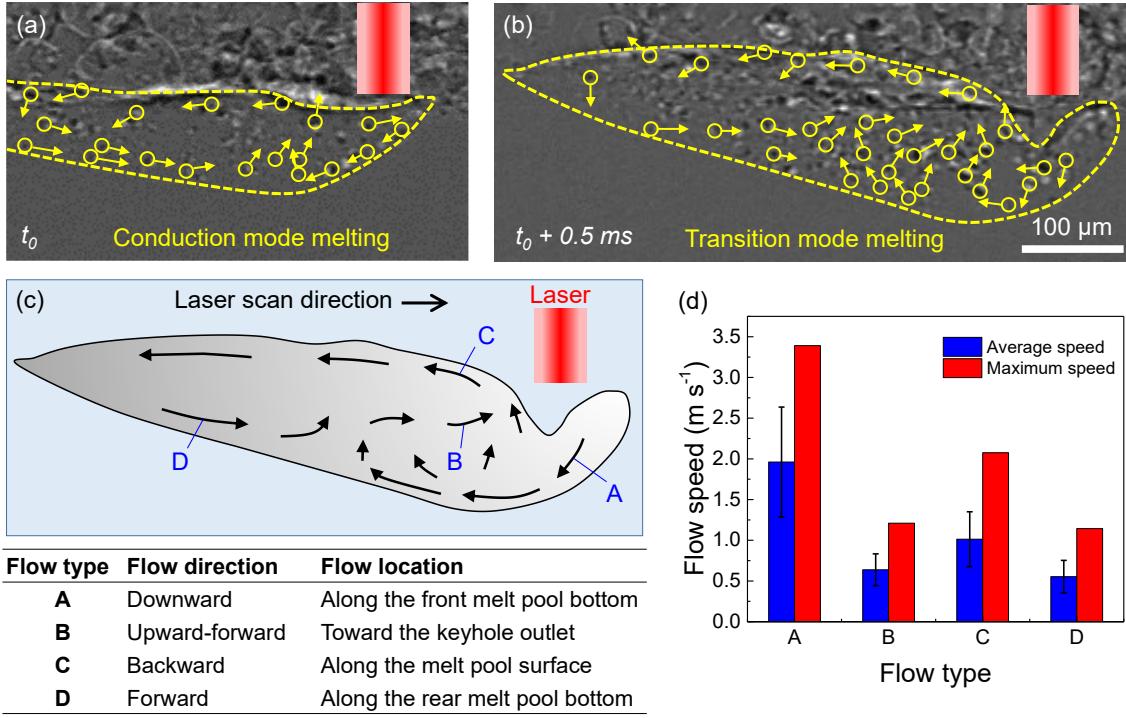
312 The instabilities unveiled above occur under a single melting mode without melting-mode
313 transition during scanning. However, a distinct type of melt flow instability can be triggered by
314 the switching of melting modes, which is a common yet often overlooked phenomenon due to the
315 difficulties to investigate/recognize by ex-situ examinations.

316 We observed a melting-mode switching event during a continuous laser scanning of a $100 \mu\text{m}$
317 thick AlSi10Mg powder on an AlSi10Mg substrate, with a constant laser power of 312 W and scan
318 speed of 0.6 m/s. Initially, as shown in Fig. 7(a), the melt pool was in conduction mode, where
319 there was no visible keyhole under the laser beam. Indicated by the motion of tracers, the melt
320 flow in the melt pool exhibited a pattern consistent with the literature [23]. However, the melting
321 mode switched into transition mode after 0.5 ms without any change in processing parameters, as

322 shown in [Fig. 7\(b\)](#). By connecting the moving directions of individual tracers, the overall flow
323 pattern in the transition-mode melt pool was mapped out and schematically illustrated in [Fig. 7\(c\)](#).
324 The front-half (keyhole-adjacent region) of the melt pool exhibited a distinct pattern from the
325 conduction-mode flow due to the intensive interruption caused by the keyhole, while the rear-half
326 of the melt pool maintained a similar pattern as the conduction-mode flow. The newly-formed
327 keyhole enhanced the laser absorption [39] and led to an increased melt pool volume. As a result,
328 the melt pool depth increased from $208 \pm 10 \mu\text{m}$ (conduction-mode) to $331 \pm 7 \mu\text{m}$ (transition-
329 mode).

330 We quantified the 2D flow speed in transition-mode melt pool by evaluating the tracers' speed
331 at four locations, as shown in [Fig. 7\(c\)](#): A—downward flow along the front melt pool bottom; B—
332 upward-forward flow toward the keyhole outlet; C—backward flow along the melt pool surface;
333 and D—forward flow along the rear melt pool bottom. [Figure 7\(d\)](#) displays the average speeds and
334 maximum speeds of the four flows. Flow-A has the highest average speed of $1.96 \pm 0.68 \text{ m/s}$ and
335 a maximum speed of 3.39 m/s . Flow-D exhibited the lowest average speed of $0.55 \pm 0.2 \text{ m/s}$ and
336 the lowest maximum speed of 1.14 m/s .

337 We did not observe a certain frequency (or period) of melting-mode switching during laser
338 scanning. As shown in [Supplementary Fig. 4](#), the melting-mode switching seems to happen
339 occasionally.



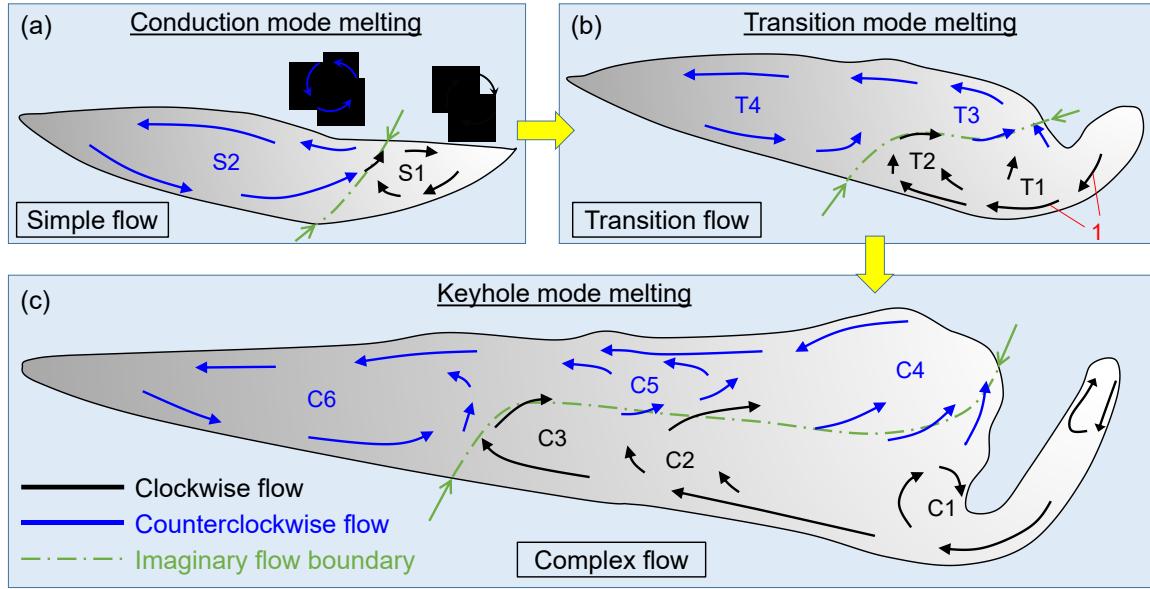
340

341 **Fig. 7. Melting mode switching induced melt flow instability.** (a, b) X-ray images showing the
342 melt flow change from conduction mode melting to transition mode melting within 0.5 ms. The
343 laser power is 312 W with a scan speed of 0.6 m/s. The material is AlSi10Mg. (c) Schematic
344 illustration of the melt flow pattern in transition mode melt pool (b). (d) Measurement of melt flow
345 speed in transition mode melt pool. Error bars represent standard deviation, $n \geq 21$ independent
346 replicates.

347 4. Discussion

348 4.1 Melt flow evolution among different melting modes

349 Although the regular melt flow patterns under conduction-mode and keyhole-mode laser
350 melting have been explicitly studied [23–25], it remains unclear how does the melt flow pattern
351 change from a simple flow (conduction-mode flow pattern, Fig. 8(a)) to a complex flow (keyhole-
352 mode flow pattern, Fig. 8(c)). Is such change arbitrary? Or is there a pattern to follow? Here, with
353 the missing-link identified in this work (transition-mode flow pattern), we clarified the complete
354 evolution path of melt flow among different melting modes, as demonstrated in Fig. 8.



355

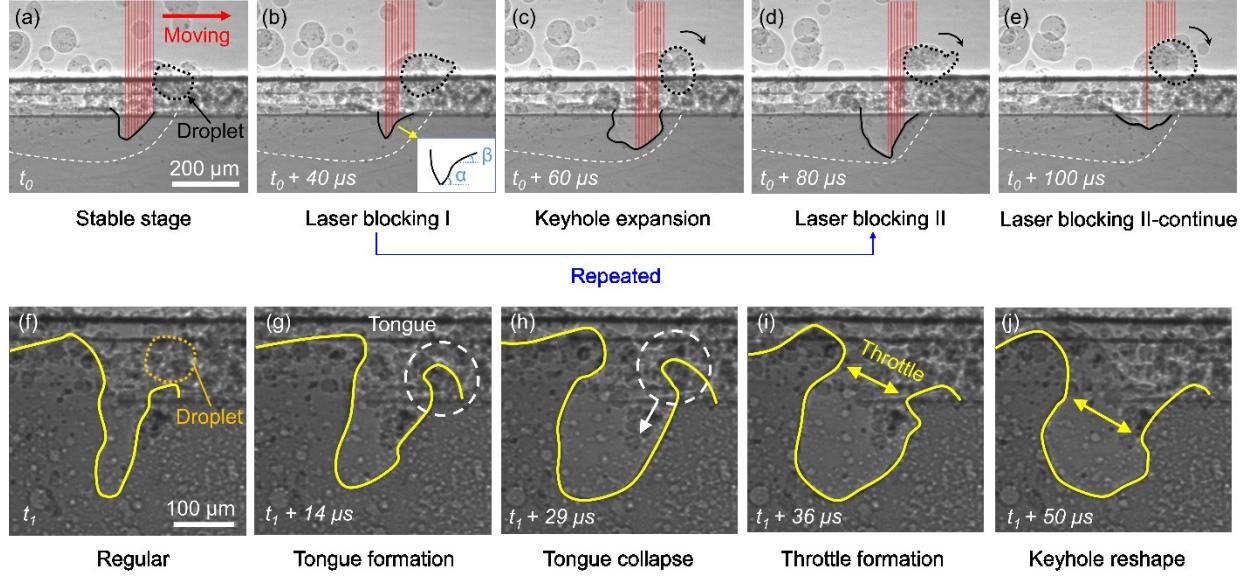
356 **Fig. 8. Melt flow evolution among different melting modes.** (a) Regular melt flow pattern in
 357 conduction mode melt pool. (b) Regular melt flow pattern in transition mode melt pool.
 358 (c) Regular melt flow pattern in keyhole mode melt pool. Black arrows mark clockwise-moving flow.
 359 Blue arrows mark counterclockwise-moving flow. Green line marks the imaginary-boundary of
 360 clockwise flow and counterclockwise flow for better interpretation.

361 In general, the complexity of the melt flow increases in scale with the melt pool size (or aspect
 362 ratio). The simple flow pattern in a conduction-mode melt pool contains two circulations—a
 363 clockwise circulation S1 and a counterclockwise circulation S2, as shown in Fig. 8(a). In
 364 transition-mode melt pool (Fig. 8(b)), the shallow keyhole exerted extra momentum to the
 365 downward flow (flow-1) along the front melt pool boundary, transporting the flow further into the
 366 body of melt pool. Compared with the clockwise circulation S1 in the conduction mode, this region
 367 was stretched into two clockwise vortices T1 and T2 in transition mode. Similarly, the
 368 counterclockwise circulation S2 was also elongated into two partial counterclockwise vortices T3
 369 and T4, as shown in Fig. 8(b). When it comes to keyhole mode, Fig. 8(c), the deep keyhole pushed
 370 the clockwise flow deeper into the melt pool. Thus, several vortices, C1, C2, and C3, formed along
 371 the path. The counterclockwise region was stretched even longer, partitioned by several
 372 counterclockwise flows C4, C5, and C6. Therefore, the melt flow patterns from conduction mode
 373 to keyhole mode gain complexity by the expansion of the clockwise region and counterclockwise
 374 region, with more vortices forming in each region.

375 **4.2 Mechanisms for significant keyhole oscillation**

376 We have identified keyhole oscillation as an important source for melt flow instabilities.
377 However, the mechanisms for causing significant keyhole oscillation are also various. Previous
378 modelling works have proposed the extra reflection of laser beam within the keyhole as a source
379 for causing keyhole fluctuations [40,41]. The unevenly distribution of laser energy on the keyhole
380 surface was also identified by multi-physics modelling to cause keyhole fluctuations [34].
381 Experimental work based on in-situ synchrotron imaging has revealed keyhole oscillations could
382 be induced by opposite flows around keyhole, or by the variation of laser absorption on
383 nonuniformly-packed powder bed [42]. It was also reported that the presence of powder could also
384 induce keyhole fluctuations [13], yet no detailed mechanisms were revealed. Here, we report two
385 new powder-based mechanisms for causing significant keyhole oscillations, as shown in [Fig. 9](#).

386 The first mechanism is laser-blocking induced keyhole oscillation, as demonstrated in [Fig. 9\(a–e\)](#). At stable stage, the laser beam will incident on the front keyhole wall ([Fig. 9\(a\)](#)). However,
387 sometimes the powder agglomerate ahead of the laser and form a large, floating droplet on powder
388 bed[9], as circled by the dashed line in [Fig. 9\(a\)](#). Once the moving laser catch up with the droplet,
389 the laser beam could be partially blocked by the droplet ([Fig. 9\(b\)](#)). The front keyhole wall under
390 the blocked-beam elevated due to the less-intensive vaporization, as shown in the inset of [Fig. 9\(b\)](#),
391 leaving a reduced inclination angle (β) of front keyhole wall as compared with the large inclination
392 angle (α) under regular laser radiation. The overall keyhole size also shrank due to the insufficient
393 laser radiation. In the next moment, [Fig. 9\(c\)](#), the localized vaporization on the droplet pushed the
394 droplet moving along the laser scanning direction and left the laser radiation area. A sudden release
395 of the laser energy to the keyhole promoted the intensive vaporization-induced recoil pressure,
396 which expanded the keyhole cavity rapidly. In this manner, the keyhole completed an oscillation
397 cycle by the laser block-unblock induced keyhole shrinkage-expansion process. The oscillation
398 frequency depends on how often the laser is blocked. For example, we captured two laser-blocking
399 events within 40 μ s, as displayed in [Fig. 9\(b–d\)](#). The oscillation amplitude depends on how much
400 of the laser energy is blocked. For example, the keyhole size only shrank when the laser is half-
401 blocked in [Fig. 9\(b\)](#), while the keyhole cavity almost vanished when the laser is nearly fully-
402 blocked in [Fig. 9\(e\)](#).



404

405 **Fig. 9. Mechanisms for causing significant keyhole oscillation.** (a-e) Laser-powder interaction
406 induced significant keyhole oscillation. The laser power is 520 W with a scan speed of 0.4 m/s.
407 The material is Al6061. (f-j) Powder incorporation induced significant keyhole oscillation. The
408 laser power is 364 W with a scan speed of 0.6 m/s. The material is AlSi10Mg.

409 The second mechanism is powder-incorporation induced keyhole oscillation, as shown in Fig.
410 9(f-j). The beginning of this event is similar to the first mechanism, where a floating droplet on
411 powder bed formed ahead of laser beam during laser scanning in Fig. 9(f). However, instead of
412 being pushed away, the droplet was captured by the front rim of the melt pool and formed a
413 “tongue”-shape protrusion (Fig. 9(g)). The tongue then collapsed into the keyhole by moving
414 downward along the front keyhole wall (Fig. 9(h)). The inclined tongue, together with the rear rim
415 of the keyhole, formed a throttle at the keyhole outlet (Fig. 9(j)), which restricted the exhaust of
416 metal vapor and also guided more laser reflection from the front keyhole wall toward the rare
417 keyhole wall [43]. As a result, the keyhole developed into a pocket shape, with an increased width
418 over three times large as the regular keyhole width. The expanded keyhole persists as long as the
419 throttle exists. However, the keyhole profile will keep being reshaped by the throttle displacement,
420 as shown in Fig. 9(j), leading to continuous disturbances to the surrounding area before the throttle
421 fades away.

422 It should be emphasized that all the observations reported in the present work are the projected
423 information on the 2D imaging plane. Although the selected 2D imaging plane does not account
424 for the out-of-plane (3D) particle movement, the tracer movement within the selected imaging

425 plane can fairly reflect the physics underlying flow instabilities. The reasons being: (1) Statistically,
426 the tracers have more tendency to move within the selected imaging plane rather than moving out
427 of plane, because the melt pool shape in LPBF is usually elongated along the laser scan direction.
428 It has been reported that the melt pool length can be several times larger than the width during
429 LPBF process [4]. (2) The melt flow patterns exhibit more complexity on the selected imaging
430 plane (perpendicular to the X-ray beam) rather than on the plane parallel to the X-ray beam [23].

431 Therefore, the instabilities we observed on 2D projection plane are valid and not affected by the
432 out-of-plane (3D) particle movements. However, there may be chances that we miss the instability
433 that happens within the plane parallel to the X-ray beam.

434 **5. Conclusion**

435 In the present work, we experimentally revealed the melt flow stabilities in laser metal additive
436 manufacturing process by in-situ high-speed high-resolution synchrotron X-ray imaging. The
437 major conclusions are drawn below:

438 (1) We identified three mechanisms as the major cause for melt flow instabilities, namely:
439 powder/droplet impact, significant keyhole oscillation, and melting-mode switching. We
440 demonstrated that these instabilities could roughen the part surface finish, break the energy
441 balance within the melt pool (by changing the instant laser absorption), and disturb the
442 solidification process at the melt pool solid-liquid interface.

443 (2) We unraveled the evolution path of melt flow pattern among different melting modes. The
444 melt pool was found to be separated into a clockwise flow region and a counterclockwise
445 flow region. The elongation of the two regions facilitated the melt pool development from
446 simple flow to complex flow.

447 (3) We explored two mechanisms for causing significant keyhole oscillation. One mechanism
448 is the laser-blocking induced keyhole oscillation, where powder droplets could occasionally
449 block the laser path and reduce the energy input to the keyhole. The other one is the powder-
450 incorporation induced keyhole oscillation, where the capturing of new particles reshapes the
451 keyhole profile.

452 The process instability mechanisms revealed in this work provide the foundation for
453 development of processing approaches to mitigate instabilities in laser metal additive

454 manufacturing processes. The melt flow dynamics revealed here are important for the
455 development and validation of high-fidelity computational models.

456

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