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Keyhole fluctuation and pore formation mechanisms during laser powder bed fusion additive manufacturing

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Abstract

Keyhole porosity is a key concern in laser powder-bed fusion (LPBF), potentially impacting component fatigue life. However, the dynamics of keyhole porosity formation, i.e., keyhole fluctuation, collapse and bubble growth and shrinkage, remain unclear. Using synchrotron X-ray imaging we reveal keyhole and bubble behaviours, quantifying their formation mechanisms. The findings support the hypotheses that: (i) keyhole porosity can initiate not only in unstable, but also transition keyhole regimes, created by high laser power-velocity conditions, causing fast radial keyhole fluctuations (~10 kHz); (ii) transition regime collapse tends to occur part way up the rear-wall; and (iii) immediately after keyhole collapse, the bubble grows as pressure equilibrates then shrinks due to metal-vapour condensation. Concurrent with condensation, hydrogen diffusion into the bubble slows the shrinkage and stabilises the bubble size. The physics revealed here can guide the development of real-time monitoring and control systems for keyhole porosity.

Laser powder-bed fusion (LPBF) additive manufacturing is being widely explored in both industry and academia^{1,2} for the production of metal parts. During LPBF, mid-power ($\sim 100 - 1000$ W) but tightly-focused (spot sizes $\sim 20 - 100$ μm) lasers are scanned across successive layers of fine metal powder at high speed ($\sim 0.05 - 4$ $\text{m}\cdot\text{s}^{-1}$), selectively melting and consolidating the powder to build a fully dense part. The typical processing-structure-property linkage for LPBF is: steep thermal gradients and high cooling rates³ ($\sim 10^4 - 10^6$ $\text{K}\cdot\text{s}^{-1}$) favouring fine, columnar grains oriented along the build direction, producing as-printed LPBF parts that exhibit increased strength, reduced ductility, and increased microstructural and mechanical property anisotropy⁴.

The laser fluence during LPBF is sufficient to vaporise the metal, generating a recoil pressure that pushes molten metal away from the laser-matter interaction zone⁵. With increasing laser fluence, the recoil pressure is large enough to open a deep, high aspect ratio vapour depression, referred to as a keyhole⁶. This is commonly used in laser welding to achieve thin and deep joints⁷. LPBF also often operates in keyhole mode melting⁶ to ensure full fusion between successive layers. Additionally, laser absorptivity increases dramatically in keyhole melting due to multiple reflections of the laser beam along keyhole⁸, opening the door for fabrication of highly reflective materials (e.g., aluminium matrix composites with $\sim 91\%$ reflectivity⁹) by LPBF, or enable a more economical laser heat source (e.g., diode laser) to be used in LPBF without sacrificing build efficiency¹⁰. However, the keyhole is subjected to axial fluctuations and radial perturbations¹¹ that are governed by the balance of energy and pressure¹²⁻¹⁴, posing a significant risk for keyhole instability^{15,16} and in some cases, collapse. Keyhole collapse often results in the formation of a bubble in the melt pool, which may get trapped by the solidification front to form a pore. Keyhole pores remaining in the final part may act as stress concentrators and sites for crack initiation and growth, making them potentially detrimental to fatigue life and other final component mechanical properties¹⁷.

Several process models^{5,12-14,18} explained the physics of keyhole pore formation during laser welding and LPBF, revealing the interactive effects of recoil pressure, surface tension, and Marangoni convection on the keyhole, and the competing influence of gravity, drag, buoyancy and thermocapillary forces on bubble motion. Recently, *in situ* synchrotron X-ray imaging¹⁹⁻²² has been applied to LPBF, capturing some

dynamics of the keyhole and keyhole pore in the sub-surface of melt pool, including: keyhole morphology evolution²³; pore formation at turn-around points during raster scanning²⁴; pore elimination by thermocapillary forces²⁵; pore migration under Marangoni-driven flow and pore coalescence^{26,27}; pores being pushed away from keyhole tip by the acoustic waves emanating from the keyhole collapse¹⁶, and pore evolution during multi-layer LPBF^{28,29}. However, the dynamics of keyhole pore formation are still not fully understood. The role of keyhole fluctuations in keyhole collapse and the evolutions of bubbles (e.g., formation, growth, shrinkage and migration) before being captured by the solidification front, are largely unexplored. For the latter, previous studies^{30,31} explored the influence of evaporation and condensation on the dynamics of water-vapour bubbles in a superheated liquid, and effect of dissolved gas diffusion on the bubble growth in casting^{32,33}, but it remains unclear how these physics extend to the LPBF process.

Here, we perform *in situ* synchrotron X-ray imaging during LPBF of a commercial aluminium alloy Al7A77 (HRL laboratory, USA), which has critical applications in aerospace, biomedical and automotive industries³⁴ and also a high laser reflectivity³⁵, presenting challenges for laser processing. We discover a transition regime (II) between the stable (I) and unstable (III) keyhole regimes in LPBF, where the keyhole morphology changes from wide and shallow in II to narrow and deep in III. Pores are also observed to form in II, mostly present at the rear keyhole wall (RKW), while keyhole porosity is more prevalent in III with pores typically forming at the keyhole bottom. Although some prior work has suggested keyhole fluctuation are largely random, we observe regular oscillations in keyhole width and depth with significant trends in fluctuation frequency across the three keyhole regimes. We find these regimes are well defined by the front keyhole wall (FKW) angle, which collapses to a single function of the normalized enthalpy product³⁶ for different materials. By comparing our bubble model with the experimental data, we find that the bubble dynamics are defined by fast initial growth induced by the pressure equalisation, followed by shrinkage due to metal-vapour condensation. Concurrent with condensation, hydrogen may diffuse into the bubble, reducing the bubble shrinkage and stabilizing the bubble size. Further, we investigate the rapid distortion of bubbles as they interact with the advancing cellular-dendrite solidification front.

Results

Keyhole collapse mechanism and related regime transitions. *In situ* and operando X-ray imaging was used to probe the keyhole collapse behaviours and keyhole pore formation mechanisms during LPBF, which was carried out using an *In Situ* and *Operando* process replicator (ISOPR, Supp. Fig S.10), as described in Methods. We systematically characterised the resulting change in keyhole shape and bubble development across a wide range of area energy densities AED^{37} , $AED = P_l/(v_l d_l)$ (P_l laser power, v_l laser scan velocity, d_l laser beam diameter), from $AED = 6$ to $17 \text{ MJ}\cdot\text{m}^{-2}$ in the keyhole melting regime⁶. We observed that the keyholes change in morphology from wide and shallow to narrow and deep (Fig. 1a, Supp. Fig S.1a and Movies 1 – 12). Simultaneously, bubbles first form at the RKW, then prevail at the bottom of keyhole once the keyhole becomes deep and narrow (Fig. 1a, Supp. Fig S.1a). Those findings indicate that the transition from a stable to unstable keyhole melting may be more nuanced than previously suggested^{16,23} (discussed in detail later). We also noticed that the FKW remained relatively smooth at an approximately constant inclination, whereas the RKW presented random wrinkles and perturbations. With increasing AED , the keyhole penetration depth increases and the inclination of the FKW become steeper (higher FKW angle θ , $\tan \theta \sim v_d/v_l^{38}$), which is attributed to higher drilling velocity³⁹ (v_d) and increased energy coupling due to multiple reflections⁸.

By supplementing our results with previous studies^{16,23,39,40,41} across a wide range of powder materials, process conditions with different LPBF replicators and beamlines, we found that the FKW angle θ collapses to an inverse tangent of the normalised enthalpy product $(\Delta H/h_m \cdot L_{th}^*)^{36}$ (Fig. 1b), where L_{th}^* is the normalised thermal diffusion length³⁶ and the normalised enthalpy $\Delta H/h_m^{6,42}$ is the ratio of ΔH , the deposited energy density⁶ (also named as specific enthalpy) and h_m , the enthalpy at melting. This relationship is derived by the governing laws of heat transfer and kinematic equilibrium, elaborated in Supp. Note 1. The agreement (Fig. 1b) between the theoretical derivation and experimental measurements, suggests that the FKW inclination during LPBF is not only controlled by the deposited energy density ΔH and the material's melting enthalpy h_m , but also affected by the thermal diffusion length L_{th} .

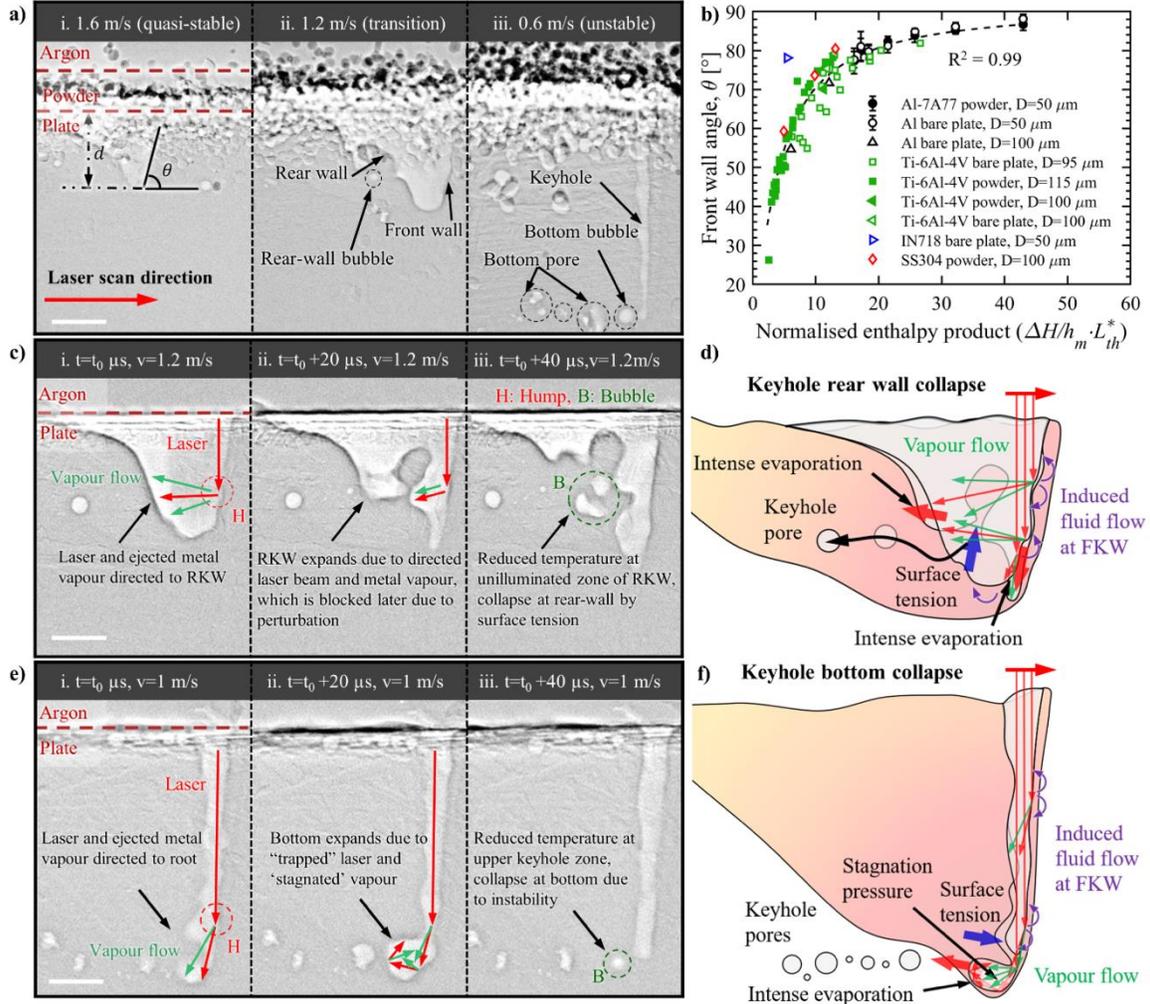


Fig. 1. Keyhole collapse mechanism and related keyhole melting regime transitions in LPBF. (a) Keyhole morphology variations across the (I) quasi-stable, (II) transition and (III) unstable keyhole regimes under different laser velocities. (b) Front keyhole wall (FKW) angle as a function of normalised enthalpy product for 9 datasets with 4 different materials. Curve fit is $\theta = \arctan(a \cdot [(\Delta H/h_m \cdot L_{th}^*) + b]) + c$ (a, b and c are constants), performed in Matlab using the Levenberg-Marquardt/least absolute residuals robust fitting algorithm. (c) Radiographs of laser melting with bare aluminium plate in (II) transition regime, showing rear wall collapse with associated illustration (d). (e) Radiographs of laser melting with bare aluminium plate in (III) unstable regime, showing keyhole bottom collapse with associated illustration (f). t_0 is the time moment that the rear keyhole wall (RKW) starts to grow. The red, blue and green arrows in (d) and (f) represent the laser beam, fluid flow and vapour flow, respectively. d and θ represent the keyhole depth and FKW angle, respectively. Laser power 500 W, laser spot diameter 50 μm . All scale bars correspond to 150 μm . The datasets of LPBF with Ti-6Al-4V are cited from Cunningham *et al.*²³ (Figs.4, S5 and S7) and Zhao *et al.*¹⁶ (Movie S1 -Movie S5) with permission by AAAS. Datasets for LPBF with Inconel 718, SS 304 and aluminium bare plate are cited from Kouraytem *et al.*³⁹, Parab *et al.*⁴⁰, and Hojjatzadeh *et al.*⁴¹, respectively.

Previous work has related the FKW to the laser fluence, but has largely neglected the role of the thermal diffusion. Cunningham *et al.*²³ reported a nonlinear relationship between the FKW angle and the power density ($2P_l/\pi d_l^2$), and this relationship changes with the laser scan velocity as well as powder materials. Gan *et al.*⁴³ found that the tangent of FKW angle is approximately proportional to the “keyhole number Ke ” ($Ke = \frac{1}{\sqrt{\pi}} \cdot \Delta H/h_m$), which is a scaled version of the normalised enthalpy. Here, we find even stronger agreement between the FKW angle and the normalised enthalpy product (Supp. Fig S.2d), rather than the normalised enthalpy (Supp. Fig S.2c). Our result builds on the work of Ye *et al.*³⁶, who first introduced the normalised enthalpy product in their scaling laws for keyhole depth (similar relations for keyhole depth measurements are shown in Supp. Fig S.2a, b). Of interest, the normalised thermal diffusion length can be expressed in terms of the Péclet number Pe , $L_{th}^* = \frac{1}{\sqrt{Pe}}$ (Methods), thus, the above function of FKW angle can be presented as $\theta \sim \text{atan} \left[\left(\frac{\Delta H}{h_m} \right) / \sqrt{Pe} \right]$, suggesting that the mode of thermal transfer (convection vs. conduction) also affects the keyhole morphology evolution during LPBF. The relationship derived here also allows for defining thresholds between different melting regimes, similar to King *et al.*⁶, who found the transition from conduction to keyhole melting occurs at a normalised enthalpy $\Delta H/h_m \approx (30 \pm 4)$ for 316L stainless steel.

Within the keyhole-melting regime, recent studies have reported a sharp transition between stable and unstable keyhole melting, typically defined by the onset of keyhole porosity^{16,23,43}. From our data, we observed that the threshold for this transition can vary significantly between alloys. For Al7A77 and Ti-6Al-4V^{16,23} (Fig.2d), we found this transition occurs at $\Delta H/h_m \cdot L_{th}^* \sim (8 \pm 3)$ or $\sim 60^\circ$ FKW angle, and $\Delta H/h_m \cdot L_{th}^* \sim (20 \pm 3)$ or $\sim 80^\circ$ front-wall angle, respectively. The larger threshold for Al7A77 is likely a combined result of its low absorptivity at ambient temperature (~ 0.15 vs. ~ 0.45), larger Brewster angle ($\sim 85^\circ$ ⁴⁴ vs. $\sim 80^\circ$, Supp. Fig S.5), and lower melting enthalpy ($h_m = 2.63 \text{ J} \cdot \text{mm}^{-3}$ vs. $6.26 \text{ J} \cdot \text{mm}^{-3}$).

In addition, we find that there can be an extended transition regime (II) between the stable (I) and unstable (III) keyhole regimes under high-power-velocity (high- PV). Pores begin to form in this transition

regime, and initiate at the RKW rather than at the bottom of the keyhole (typical in III), which was also observed during laser welding of aluminium alloys¹⁵ and low carbon steel⁴⁵, as well as LPBF of Ti-6Al-4V⁴¹. For similar *AED*, we found this transition regime becomes sharper with decreasing laser power and scan speed ($P_l = 500\text{ W}$, $v_l = 1.4\text{ m/s}$, Supp. Fig S. 3a, b; $P_l = 200\text{ W}$, $v_l = 0.6\text{ m/s}$, Supp. Fig S. 3c, d), in agreement with Zhao *et al.*¹⁶. We speculate that the (II) transition regime is induced by the high-*PV* combination under large *AED*, which enlarges the melt pool and vapour depression zone, leading to a relatively wider transition from the stable (I) to the unstable (III) keyhole regime. For the laser spot size and alloy used in this study, a high-*PV* with large *AED* is defined as $P_l = 500\text{ W}$, $v_l = 1.2\text{ m/s}$, $AED \geq 7\text{ MJ}\cdot\text{m}^{-2}$. Considering that the general *AED* in LPBF is around $10\text{ MJ}\cdot\text{m}^{-2}$ based on reference¹⁴, we speculate that this transition regime (II) would become popular when high-*PV* processing is required to achieve large build rate in LPBF.

To further investigate the different keyhole collapse mechanisms in II and III, we compared the keyhole dynamics (Fig.1c, Fig.1e and Supp. Movies 1 – 12). “Humps” regularly form on the FKW due to the dependence of laser absorption on angle of incidence (Fresnel absorption⁴⁶, Supp. Note 2), which becomes especially pronounced around the Brewster angle⁴⁶ (above which, absorptance falls off dramatically, Supp. Fig S.5). In II (Fig.1c-i, $AED = 8\text{ MJ}\cdot\text{m}^{-2}$, FKW angle $81.2 \pm 1.7^\circ$), these humps tend to reflect the laser beam and the vapour flow towards the RKW. This leads to intensive evaporation and recoil pressure on the RKW and builds up a stagnation pressure¹², correspondingly deforming and expanding the RKW (Fig.1c-ii). Generally, the combined recoil and stagnation pressure balances the surface tension acting on the free surface of the RKW, holding the overhanging RKW from collapse¹⁴. However, should the reflected laser beam and vapour flow be blocked or redirected by a perturbation of the keyhole (Fig.1c-ii), the surface temperature of the unilluminated RKW will quickly decrease. As the temperature decreases, surface tension increases linearly⁵, overcoming the recoil pressure which decreases exponentially⁵, causing a RKW collapse. We observed that this collapse can sometimes lead to the formation of bubbles from the RKW, approximately at the half-depth of keyhole (Fig.1c-iii), followed by the temporary formation of a deep, high aspect ratio depression. The melt flow at the middle of the pool half way up the

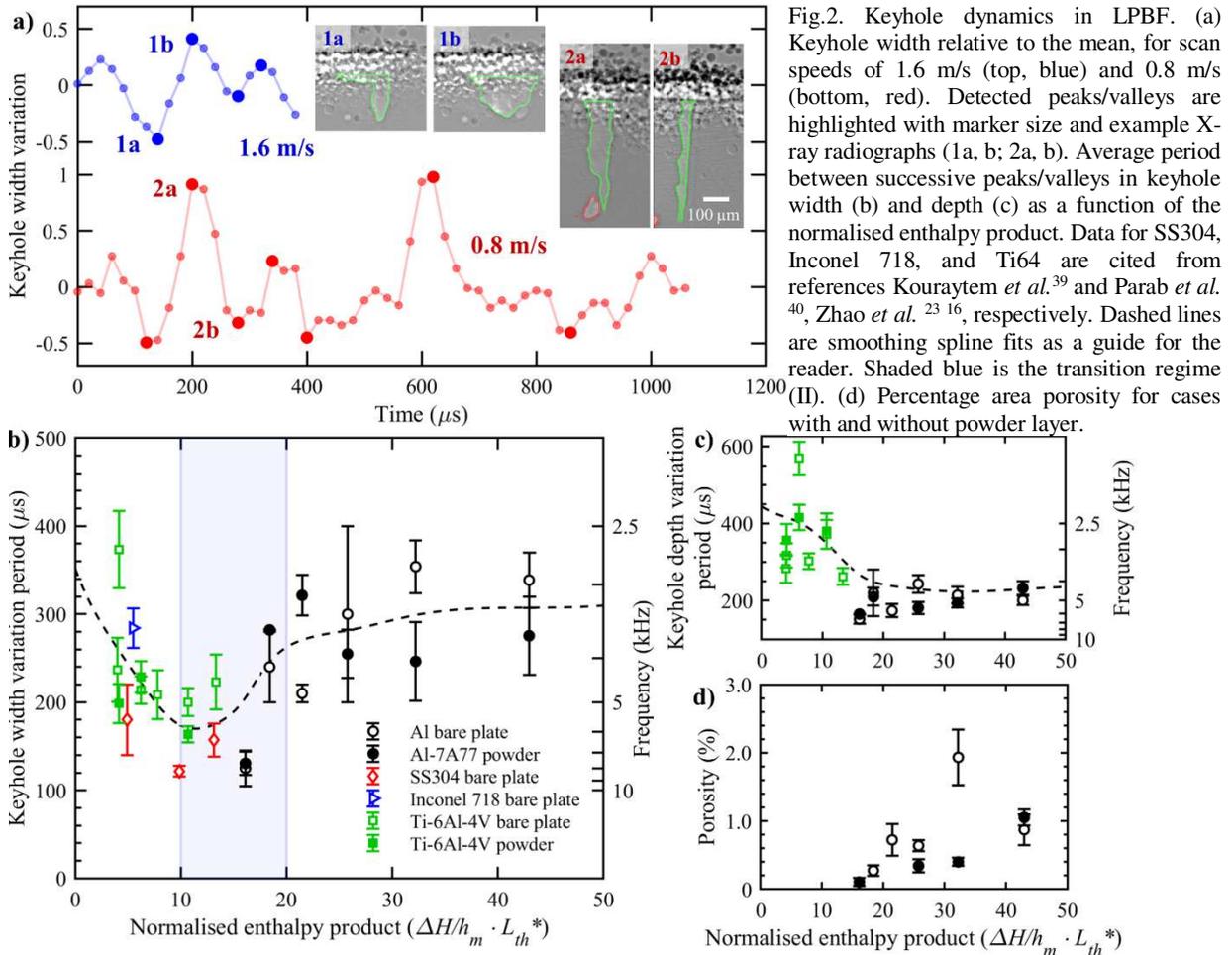
RKW is still strong⁴⁷, as a result of the Marangoni-driven flow, propelling the pore towards the rear of the pool, as discussed in detail later.

In the unstable regime (III) (Fig. 1e-i, $AED = 10 \text{ MJ}\cdot\text{m}^{-2}$, FKW angle $84.8 \pm 0.8^\circ$), a narrow, deep keyhole forms, and humps on the FKW direct metal vapour and reflected laser beams to the bottom of the keyhole. Intense evaporation and recoil pressure at the keyhole bottom can be further amplified by the rapid formation of a vapour cavity (“J-shaped” keyhole, Fig. 1e-ii), which traps reflected laser light and metal vapour, increasing the number and density of multiple reflections^{8,18} and building up a significant stagnation pressure¹². With energy concentrated in this cavity, the keyhole is prone to capillary instability and may sometimes collapse, pinching off a cavity to form a vapour filled bubble (Fig. 1e-iii) and leads to a sharp decrease in keyhole depth. This is similar to, but not the same as the “spiking” as initially named in laser welding¹¹. Spiking is also prevalent in LPBF but at turn-around points in raster scan patterns due to the finite acceleration of the laser beam, near-zero instantaneous scan velocity, and resulting pores at the “root” of keyhole²⁴. While a small number of the bubbles we observed were re-captured by the expanding keyhole (Supp. Fig S.1b), most were captured almost instantaneously by the advancing solidification front at the bottom of the keyhole to form pores.

Keyhole radial and axial fluctuation and keyhole porosity. To quantify the keyhole and bubble dynamics, we built an image processing pipeline (Methods) to extract the keyhole depth and width from *in situ* X-ray radiographs (Fig.2). This was carried out for both our own study of LPBF with and without Al7A77 powder (Movies 1 – 8 and 10 – 12), as well as a number of previous synchrotron X-ray studies^{23,39,40} across different powder materials, process conditions, LPBF replicators, and beamlines (e.g., Parab *et al.*⁴⁰). Note that the keyhole width is extracted as the median width along the whole keyhole depth.

Fig.2a shows the regular fluctuations in the keyhole width across different keyhole melting regimes (transition II, blue; unstable III, red). Similar, if not more regular, fluctuations were also observed without powder (Supp. Fig S.4). To further quantify these fluctuations, we calculated the average peak-to-peak period (Methods) and found the corresponding frequencies of keyhole depth and width fluctuations range

from ~ 2.5 kHz to ~ 10 kHz, in agreement with previous acoustic, optical and radiometric measurements^{8,48,49}. We also found significant trends in the keyhole width and depth fluctuations across different keyhole regimes (Fig.2b): starting in I ($\Delta H/h_m \cdot L_{th}^* < 10$), the keyhole width fluctuations first increase in frequency, reach the highest in II ($10 < \Delta H/h_m \cdot L_{th}^* < 20$), and then slightly decrease in frequency in III ($\Delta H/h_m \cdot L_{th}^* > 20$). Similar patterns for the keyhole depth fluctuation are shown in Fig.2c, which increases in frequency from I to II, and then remains high in III.



The keyhole width and depth fluctuation trends are consistent with the keyhole collapse mechanisms discussed above. In II, the high frequency hump formation and subsequent migration down the FKW (Supp. Fig S.13) can cause an open, wide vapour depression to temporarily collapse into a deeper, higher aspect ratio keyhole, with a significant decrease in keyhole width and increase in keyhole depth (although sometimes less significant), boosting the fluctuation frequency of keyhole. In III, relatively higher

oscillation frequencies for the depth vs. the width also agrees with the discussion of Fig.1, corresponding to bubbles being pinched off at the keyhole bottom, followed by a sharp decrease in keyhole depth. As shown in Fig.2d, keyhole pores begin to form in II and increase in frequency through III. Comparing the final depth of pores relative to the substrate with the average keyhole depth (Supp. Fig S.6) corroborates that bubbles initiate both at the RKW in II and at the keyhole bottom in III.

Prior studies^{23,16} reported larger keyhole fluctuations with powder compared to bare substrate. Zhao *et al.*¹⁶ hypothesised that this phenomenon is induced by the momentary interaction between particle spatter and the laser beam¹³, which shades the laser illumination and reduces recoil pressure, correspondingly increasing keyhole fluctuation. Here, by comparing the fluctuation frequency of keyhole width (Fig.2b), depth (Fig.2c), and also the tracked bubble numbers at per unit track length (Supp. Fig S.7) with and without powder, we observed limited differences between the powder and bare plate samples. We hypothesise that the shadowing effect of particle spatter on the laser beam is less significance when a high laser power and a thin powder layer thickness are applied (for the laser spot size and alloy used in this study, a high laser power and a thin layer thickness is defined as ≥ 500 W and ≤ 30 μm , respectively), which is consistent with the finding reported by Khairallah *et al.*¹³. Khairallah *et al.* found that there exists a power threshold beyond which the particle spatter expulsion mechanism is activated and could vaporise the spatter quickly, inversely, inducing pores due to laser shadowing with rapid cooling.

Keyhole-induced bubble lifetime dynamics in LPBF. Using our image processing pipeline (e.g., Kalman filter tracking⁵⁰), we traced the evolutions of the keyhole-induced bubbles and extracted their centroids and equivalent diameters over their lifetime, starting after a bubble is pinched off from the keyhole and ending when the bubble is fully captured by the solidification front (see examples in Fig.3a,b, $AED = 10 \text{ MJ} \cdot \text{m}^{-2}$, Movies 7, 8; Supp. Fig S.8a, b, $AED = 17 \text{ MJ} \cdot \text{m}^{-2}$, Movies 1 – 4). We observed that bubbles evolve through three main stages, both with and without the presence of a powder layer:

- (1) bubbles rapidly grow immediately after they are pinched off from the keyhole (Fig.3a, b-i, ii, iii), thought to be due to pressure equalisation; then

- (2) the bubbles shrink while migrating towards the rear side of melt pool (Fig.3a, b-iv), hypothesised to be caused by condensation of the metal vapour in them, competing with diffusion of hydrogen into the bubbles; and finally
- (3) they are captured by the advancing solidification front (Fig.3a, b-vi).

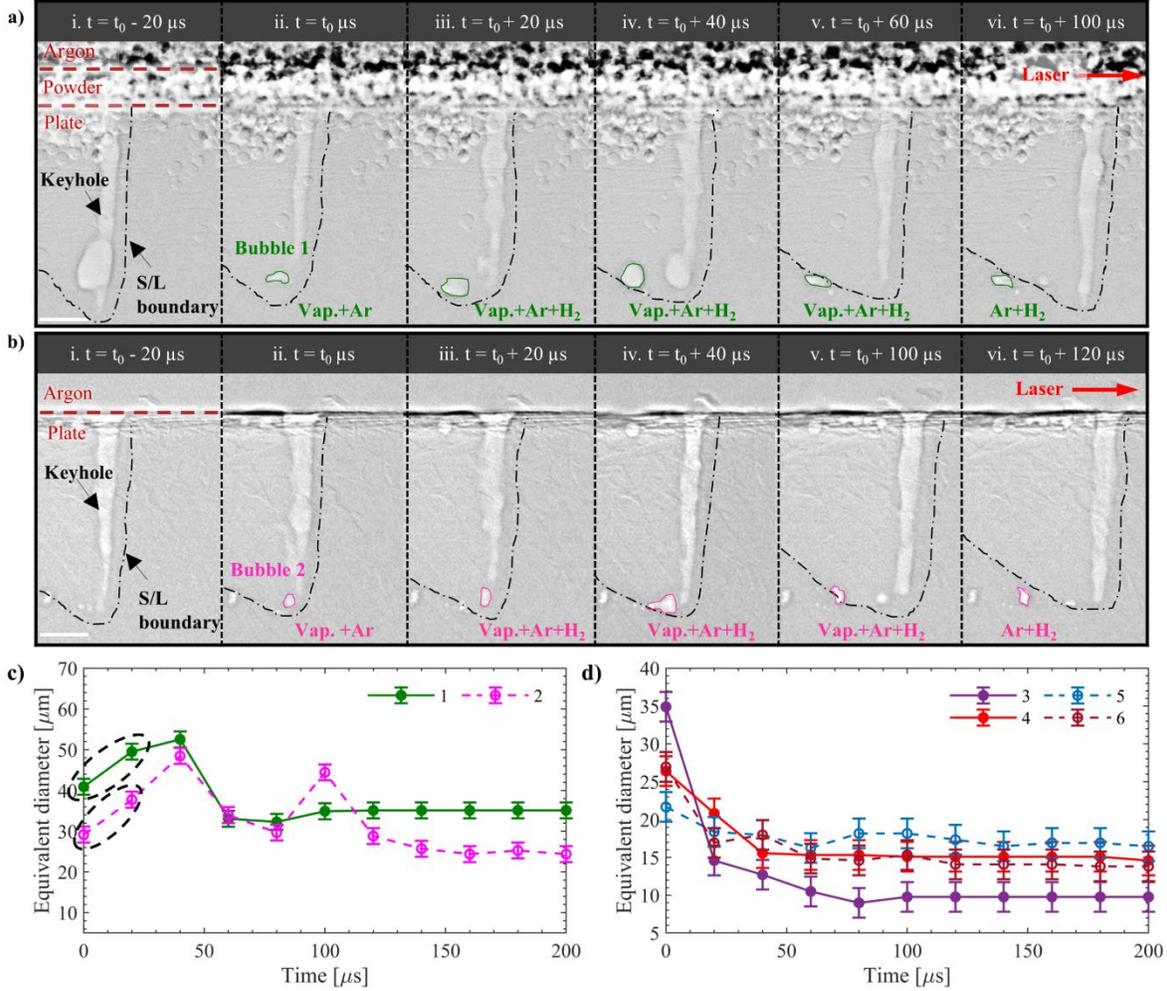


Fig.3. Keyhole bubble lifetime dynamics during LPBF. Laser velocity 1 m/s and laser power 500 W. (a) and (b) are radiographs with Al7A77 powder and bare aluminium plate, respectively. (c) and (d) show the equivalent diameter changes of some tracked bubbles in LPBF with (solid line) and without (dash line) Al7A77 powder. The equivalent diameter is calculated as $\sqrt{6A/\pi}$, A is the bubble area measured from X-ray image. Note the bubble size error is calculated as ± 2 pixels (1.96 $\mu\text{m}/\text{pixel}$), equivalent to the segmentation uncertainty. The total tracked bubble numbers are 5 and 8 for the powder and bare plate cases (Supp. Fig S.9a), respectively, using a criterion where the minimum number of frames that a bubble is identified is 6 (Methods). The time t_0 is set to the moment a bubble is first identified. The black dashed circles show initial bubble growth. The interested keyhole pores

shown in (a) and (b) are marked by green and pink colours, which are also shown with same colours in (c) and (d), respectively. Vap.: Vapour; Ar: Argon; H₂: Hydrogen. All scale bars correspond to 100 μm .

Other occasional bubble dynamics, i.e. re-captured by the keyhole, splitting and coalescence were also observed in our LPBF experiments (Supp. Movies 1 – 12). In stage (1), since the bubble was just pinched off from the keyhole, the bubble inner pressure p_i is expected to be similar to the keyhole bottom recoil pressure ($\sim 10^5 - 10^6 \text{ Pa}$ ^{14,39}), which is generally larger than the ambient pressure p_a ($\sim 1 \times 10^5 \text{ Pa}$). This pressure difference then drives bubble growth according to the ideal gas law⁵¹ ($p = nRT/V$), where the volume, V , must increase to accommodate the reduction in pressure p from p_i to p_a (Note, n is the molar number of gas, R the universal gas constant, and T the temperature). Simultaneously, as the surrounding liquid metal cools the bubble, the superheated metal-vapour inside the bubble will condense, reducing n , and hence decreasing the bubble volume V , but at a slower rate than the pressure equalisation (discussed in detail later). This is also known as the bubble contraction mechanism in laser welding^{15,52}.

In stage (2) bubbles shrink while migrating towards the rear side of melt pool, we observed that the bubble shrinkage undergoes a marked slowdown at the later stage of condensation (e.g., bubble 3 from 40 μs to 120 μs in Fig.3d), and the bubble size then get stabilized. We speculate the slowdown shrinkage and bubble size stabilization are caused by the hydrogen diffusion³². The presence of hydrogen in keyhole pores was observed by Matsunawa *et al.*¹⁵, who measured $\sim 3 - 12\%$ hydrogen content in pores formed during laser welding of aluminium alloy using mass spectrometry. Hydrogen is expected to be present in both the virgin substrate and powder particles. During LPBF, the melt at the advancing solidification front can then become supersaturated with hydrogen, driving hydrogen diffusion from the melt into the bubble^{28,30} and it is several orders faster than the diffusion of other atoms⁵³.

In stage (3) as bubbles reach the solid/liquid interface before being fully captured by the solidification front, we observed that the bubbles experience some sudden, apparent bursts of growth and shrinkage (e.g., bubble 2 at 80 – 120 μs in Fig.3c, Supp. Fig S.14a, b). This phenomenon may be explained by the interaction of the bubble with the rapidly growing microstructure^{54,55} (e.g., the bubble is pierced through, or perhaps distorted from spherical, by the growing columnar dendrite), elucidated in Supp. Note 3.

Based on the above findings, from the initiation of a bubble until it gets frozen as a pore, its composition will initially be a combination of metal vapour and shielding gas argon (Ar), which is driven into the keyhole via the Bernoulli effect⁵⁶. The metal vapour will condense, leaving the Ar, and reducing the pore size. Simultaneously some hydrogen (H₂) will diffuse in, slowing the bubble shrinkage. These stages are highlighted by the tracked bubble colours in Fig.3a, b. Note that the argon can be treated as insoluble in molten aluminium⁵⁷, which is expected to be the major content left in the frozen pore.

To verify our hypothesis in above discussion, we combined the Rayleigh-Plesset equation⁵⁸, bubble condensation model from Florschuetz and Chao⁵⁹, and the ideal gas law⁵¹ to build a united bubble model (Methods) while considering pressure-driven growth, vapour condensation and hydrogen diffusion. We compared the modelled results with experimental measurements under different keyhole melting regimes (II, III), shown in Fig.4, where we presented the tracked keyhole and keyhole pore transient trajectories at 0.8 m/s and 1.2 m/s laser velocities (Fig.4 a, b) based on the X-ray images (Supp. Movies 5, 6, 10, 11), corresponding to II ($AED = 12.5 \text{ MJ} \cdot \text{m}^{-2}$) and III ($AED = 8 \text{ MJ} \cdot \text{m}^{-2}$).

As seen from Fig.4c, d, immediately after forming the bubble experiences explosive growth, which plateaus after about $\sim 3 - 5 \mu\text{s}$ (Supp. Table S. 3). The bubble then begins to shrink at a slower rate, and eventually get stabilized ($\sim 50 - 150 \mu\text{s}$), being consistent with the experimental measurements (Note that deviations may be caused by the effect of surface tension and fluid flow that are not included in the built model). We hypothesise that the explosive growth is caused by the pressure equalisation, with the bubble volume increasing like t^3 (see Methods). The decline and plateau in bubble growth is explained by the decrease in driving force ($p_i - p_a$) with increasing bubble volume, based on the ideal gas law. We also speculate that the bubble shrinkage rate is slower than the bubble growth rate, proportional to $t^{-1/2}$ and t , respectively (see Methods). Additionally, we hypothesise that the bubble shrinkage rate can be further slowed by the diffusion of hydrogen. The modelling results (Fig.4 c, d) show that accounting for the hydrogen diffusion is necessary to explain the latter stage of bubble size stabilisation and this leads to good agreement with the tracked bubble equivalent diameter. Note that our temporal resolution of X-ray

imaging ($20 \mu\text{s}$) is insufficient to capture the whole process of bubble growth after being pinched off from keyhole. With a calculated growth time on the order of μs , it is likely that we often fail to capture this growth, explaining why immediate bubble shrinkage is observed more frequently than initial bubble growth then shrinkage in our X-ray imaging (Supp. Fig S.9).

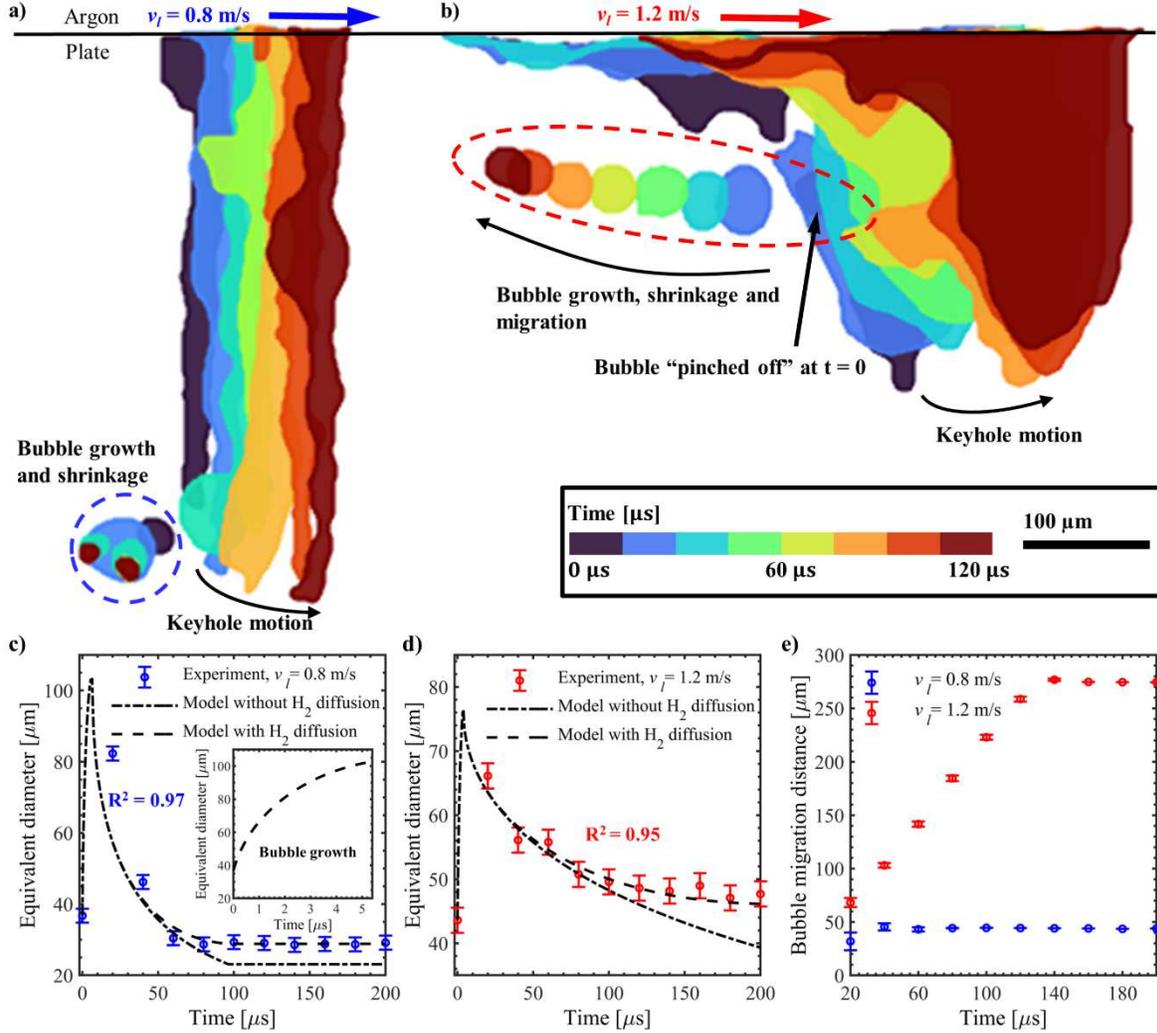


Fig.4. Tracking and modelling of keyhole induced bubble dynamics. Colour map tracking for keyhole and bubble under low (a) and high (b) laser velocities, corresponding to regimes (III) and (II), respectively. Comparing the modelled bubble size variations with *in situ* X-ray measurements at low (c) and high (d) laser velocities. The equivalent diameter is calculated as $\sqrt{6A/\pi}$, A is the bubble area measured from X-ray image by the processing algorithms listed in Methods. The bubble size error is calculated as ± 2 pixels ($1.96 \mu\text{m}/\text{pixel}$), equivalent to the segmentation uncertainty. Note, the bubble shown in (c) split into two small ones in the later stage, where the equivalent diameter is estimated based on their sum area. The temporal resolution of X-ray imaging ($20 \mu\text{s}$) is insufficient to capture the whole process of bubble growth, therefore, we are unable to get enough data and fully verify

the bubble growth model. (e) Bubble migration distance compared to their initial formed location. The bubble migration distance error is calculated based on the bubble motion with instantaneous speeds (0 – 5 m/s) during the finite camera exposure time (2.5 μ s). Low laser velocity 0.8 m/s, high velocity 1.2 m/s, laser power 500 W.

The bubbles migrated a larger distance in II with a tendency of going backward and upward, when initiated at the RKW, than in III (Fig.4e.), when initiated at the bottom of the keyhole, remaining almost stationary. This is due to two effects (i) the proximity of the solidification front to the bottom of the keyhole, bubbles having insufficient time to move upwards in the melt pool¹⁸; and (ii) the melt flow velocity and flow pattern are location dependent across the melt pool⁴⁷, and the induced drag force is local flow velocity dependent. The flow velocity is higher in the middle of the pool, hence when a bubble is formed half way along the RKW, it quickly flows backwards to the rear of melt pool. At the bottom of the pool near the solid-liquid interface, the flow is low, hence bubbles detaching from the very bottom of keyhole remain nearly stationary until captured by the solidification front and turn to pores. The bubble in Fig.4a (regime III) has an average velocity of 1.0 ± 0.5 m/s (measured over the first 60 μ s), while the bubble in Fig.4b (regime II) moves at 2.4 ± 0.7 m/s. In regime III with high *AED*, the pore migration speed of 1.0 m/s agrees well with our previous measurements of the Marangoni flow speed with tungsten carbide particles (0.97 m/s under the same *AED*)⁶⁰, while we assume that a strong viscous drag force is responsible for the higher initial speed of bubbles initiated at the RKW in regime II at lower *AED*.

Discussion

In summary, this manuscript reveals the lifetime dynamics of keyhole pore (growth, shrinkage, migration), introducing a new threshold, the normalized enthalpy product, to reveal and elucidate different keyhole pore generation mechanisms and their corresponding keyhole melting regimes in LPBF. Our findings on keyhole fluctuation and bubble dynamics provide critical guidance (e.g., bubble growth/shrinkage rate, pore location and size) to achieve *in situ* pore elimination by remelting^{25,61} upon the dual-laser LPBF machines⁶² or hybrid LPBF⁶³, and pore suppression via real-time control of keyhole dynamics (e.g., beam oscillation⁶⁴) in broad high-energy-beam processing techniques (e.g., electron beam melting⁶⁵, keyhole laser welding⁶⁴ and laser drilling⁶⁶).

In this study, we combined our own *in situ* synchrotron X-ray imaging results from LPBF of aluminium alloy (Al7A77) with recent studies of other key additive manufacturing alloys (e.g., Ti-6Al-4V, Inconel 718, SS 304) and drew the following conclusions:

- A transition regime (II) between the stable (I) and unstable (III) keyhole regimes has been discovered based on the evolutions of keyhole morphology and bubble formation. This transition regime II is pronounced for high-*PV* combinations under large *AED* ($AED \geq 7 \text{ MJ}\cdot\text{m}^{-2}$), and features the vapor depression transforms from wide and shallow to narrow and deep, concurrently, keyhole collapses and induce pores at mid-rear wall in II, as opposed to the bottom of the keyhole in III.
- Significant trends in keyhole fluctuation frequency (radial and axial) were observed across the different keyhole regimes, where the fastest fluctuation occurring in the transition regime (II) at ~10 kHz.
- A material, machine and process condition agnostic relationship was developed for the FKW angle, which collapses to a single function of the normalized enthalpy product, providing a non-dimensional threshold for predicting the three keyhole regime transitions and the onset of keyhole porosity for different alloys and processing conditions (e.g., variant laser spot sizes, powers, velocities).
- Keyhole pore formation process, including the lifetime dynamics of the vapour bubble in the melt pool, were revealed by the X-ray imaging and were summarized in three stages: (1) fast pressure-driven growth, (2) shrinkage by metal vapour condensation and shrinkage inhibition by hydrogen diffusion, and (3) interaction with growing microstructure (e.g., cellular-dendrites) and capture by the advancing solidification front.
- A model of bubble growth and shrinkage was proposed, including the physics of pressure-driven growth, vapour condensation and hydrogen diffusion. This model was found to be consistent with the experimental data, supporting our hypotheses: (i) the explosive bubble growth at the early

lifetime of stage (1) is mainly a pressure-driven process, where the bubble volume expands like $\sim t^3$; (ii) the hydrogen diffusion is sufficiently high to stabilize the bubble size at the later stage of condensation in stage (2).

Methods

LPBF Replicator (ISOPR) system and processing conditions. *In situ* synchrotron X-ray imaging was performed to probe keyhole and keyhole bubble dynamics during LPBF at the Argonne National Laboratory’s Advanced Photon Source (APS) over the ISOPR (Supp. Methods, Fig S.10). The ISOPR, custom-designed to accommodate the synchrotron X-ray imaging of the LPBF process, mainly includes a continuous wave Ytterbium-doped fibre laser (IPG YLR-500-AC, USA) with a wavelength of 1070 ± 10 nm and maximum power of 520W, an X–Y galvanometer scanner (intelliSCANde 30, SCANLAB GmbH, Germany), an environmental chamber and a sample holder positioned at the centre of the chamber. During the experiment, the chamber was filled with argon gas at a pressure of +10 kPa to reduce oxidation. The keyhole and keyhole pores were imaged at high spatial ($1.96 \mu\text{m}$) and temporal (frame rate 50 kHz) resolutions with a FASTCAM SA-Z 2100K (Photron, USA) camera by converting the attenuated x-ray beam to optically visible light using a $100\mu\text{m}$ thick LuAg:Ce scintillator. The resultant field of view was 512 pixels (1mm) in width by 680 pixels (1.33 mm) in height. The commercially Al7A77 powder (HRL Laboratory, USA) with a particle size range of 15 – $45\mu\text{m}$, and pure aluminium (Goodfellow, UK) plate with purity of 99.99% that sandwiched between two 1 mm thickness glassy carbon plates (HTW, Germany), were used in this study with the process parameters shown in Table 1. Thermophysical properties of the materials are shown in Supp. Table S.1.

Table 1. Process parameters for the LPBF experiments

Parameters	Values	Parameters	Values
Laser velocity [m/s]	0.6, 0.8, 1, 1.2, 1.4, 1.6	Laser power [W]	200, 500
Track length [mm]	5	Laser spot size [μm]	50
Layer thickness [μm]	30	Aluminium substrate size [mm]	$46 \times 17 \times 0.5$

Image and data processing pipeline - denoising, segmentation, feature extraction and tracking. The major steps of developed image processing pipeline are shown in Supp. Methods, Fig S.11 with the following steps:

- i. Flat field correction, subtracting the offset background, followed by a 2D Gaussian filtering (Supp. Fig S.11a, b). Note that the flat field correction used a general equation²⁶: $FFC = (I_0 - Flat_{avg}) / (Flat_{avg} - Dark_{avg})$, where FFC, I_0 , $Flat_{avg}$ and $Dark_{avg}$ represent the flat field corrected image, raw image, average of 100 flat filed images and average of 100 dark field images, respectively.
- ii. Initial image segmentation (Supp. Fig S.11c) with K-means clustering algorithm⁶⁷. The segmentation uncertainty is around ± 2 pixels ($1.96 \mu\text{m}/\text{pixel}$).
- iii. Frame stack integration (Supp. Fig S.11d) over the time and applying volume threshold for noise reduction.
- iv. The final segmented keyhole and keyhole bubble were achieved by applying (iii) over the whole radiograph stack. Examples are shown in Fig S.12a and Supp. Movies, 2, 4, 6, 8, 11, where the keyhole and keyhole bubble were marked with green and red colours, respectively.
- v. Kalman filter tracking algorithm⁵⁰ was developed based on the segmented keyhole bubble in step (iv). The minimum number of frames that a bubble is identified was set as 6 for being considered as an effective track. The bubbles that were spotted less than 6 frames were mainly recaptured by the successive keyhole and disappear.

Note that the segmentation algorithm developed in step (ii) fails in very limited cases due to the background fluctuations (e.g., the particle randomly fell into the sandwiched gap between glassy carbon plate and aluminium plate). Here we also developed a supervised machine leaning (random decision forests) model to achieve the segmentation of keyhole and keyhole bubble (Fig S.12b), but found that the machine leaning model fails more often during segmentation, possibly induced by the non-perfect ground-truth labelling as the X-ray frames may have different background fluctuations. The Kalman tracking algorithm developed in step (v) also fails occasionally due to the split or coalescence of bubbles, as well

as the background fluctuations. But those segmentation and tracking failure do not significantly affect the quantified keyhole and keyhole bubble dynamics.

Peak-to-peak period for keyhole width/depth. Matlab's built-in findpeaks function⁶⁸ was used to extract peaks and valleys from the keyhole width/depth signatures. To mitigate the effect of outliers and noise, the signatures were pre-processed using a moving mean filter with a width of 3. A linear fit of the data was also subtracted to remove slow changes in the width/depth. To remain safely below the Nyquist frequency (25 kHz), the minimum peak distance was set to 5 (100 μ s), acting as a simple low pass filter with cut-off frequency of 10 kHz. The minimum peak prominence was set to 1 standard deviation of the keyhole width/depth data set.

Bubble growth model with condensation and hydrogen diffusion. In the LPBF process, a rigorous modelling of the bubble dynamics while coupling the meso-nanosecond multi-physics of the process remains a challenge. Here, we assume that the contents, temperature and pressure within the bubble are homogeneous, bubble keeps spherical and be at rest relative to the incompressible melt pool flow. We take the bubble equivalent radius that the bubble is first identified as the bubble initial radius r_{b0} at the moment $t = 0$. In stage (1), considering the very short time of bubble growth, we suppose that there is negligible mass or thermal transfer over the bubble interface (assume no condensation and gas diffusion). Accordingly, the instantaneous bubble radius $r_b(t)$ at time t can be described by the Rayleigh-Plesset equation⁵⁸,

$$r_b \cdot \frac{\partial}{\partial t} \left(\frac{\partial r_b}{\partial t} \right) + \frac{3}{2} \left(\frac{\partial r_b}{\partial t} \right)^2 = \frac{1}{\rho_l} \left(p_i - p_a - \frac{2\sigma}{r_b} - 4\mu_l \cdot \frac{\partial r_b}{\partial t} \right) \quad (1)$$

where ρ_l is the density at melting temperature T_l , p_i the bubble inner pressure, p_a the ambient pressure, σ the surface tension and μ_l is the viscosity.

Since the surface tension and the viscosity terms ($\sim 10^4$ Pa) are both negligible compared to the pressure difference $p_i - p_a$ ($\sim 10^5 - 10^6$ Pa)^{14,39}, which can be omitted in the above Rayleigh-Plesset equation. Accordingly, the bubble growth rate can then be approximated as⁵⁸,

$$\frac{\partial r_b}{\partial t} \rightarrow \left(\frac{2}{3} \cdot \frac{p_i - p_a}{\rho_l} \right)^{\frac{1}{2}}, \quad r_b(t) \gg r_b(0) \quad (2)$$

This derived bubble growth rate suggests that, at the early time of stage (1) with maximum pressure difference, the bubble grows with a function of time t , while the bubble volume expands like t^3 . The initial bubble inner pressure $p_i(0)$ may be approximated as the recoil pressure p_{recoil} , $p_i(0) \approx p_{recoil}$, where the recoil pressure is a function of temperature T based on Anisimov's evaporation model^{5,14},

$$p_{recoil} = 0.54p_a \exp \left[\frac{\lambda}{K_B} \left(\frac{1}{T} - \frac{1}{T_v} \right) \right] \quad (3)$$

where $\lambda = 293.4 \text{ kJ} \cdot \text{mol}^{-1}$ is the latent heat of evaporation per atom of aluminium, $K_B = 8.314 \times 10^{-3} \text{ kJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ the Boltzmann constant, T the keyhole surface temperature, $T_v = 2753.15 \text{ K}$ is the evaporation temperature of aluminium. By using the 2D moving heat source model⁶, the keyhole surface temperature is approximated by the melt pool peak temperature,

$$T = \frac{\sqrt{2}\beta I r_l}{k_l \sqrt{\pi}} \tan^{-1} \sqrt{\frac{2a_l}{v_l r_l}} \quad (4)$$

where β is the laser absorptivity (depends on angle of incidence, see Supp. Fig S.5), r_l the laser beam radius, I the laser intensity (approximated as $I = P_l / (\pi r_l^2)$, P_l the laser power), a_l the liquidus thermal diffusivity, $k_l = \rho_l \cdot c_l \cdot a_l$ (ρ_l the liquidus density, c_l the liquidus heat capacity) the liquidus thermal conductivity and v_l the laser scan velocity.

By combing the above Equations (2 – 4) and the ideal gas law⁵¹ ($p = nRT/V$, p the pressure, n the molar number of gas, R the universal gas constant, $V = \frac{4}{3}\pi r_b^3$ the bubble volume), the transient bubble size in stage (1) can be calculated as $r_b(t) = r_{b0} + \Delta r_{b1}(t)$, $t \leq t_1$, where $\Delta r_{b1}(t)$ (solved from simultaneous Equations 2 – 4 and ideal gas law) is the pressure-driven bubble radius increment at time t , and t_1 is the pressure-driven bubble growth time. Note that t_1 is calculated as the time that the pressure difference $p_i - p_a$ reduces to a percentage threshold of p_a (we used 5% and defined as $p_i(t_1) - p_a \leq 0.05p_a$).

In the condensation dominated regime of stage (2), Florschuetz and Chao⁵⁹ built the following relation between the bubble instantaneous radius $r_{b2}(t)$ and the radius at the beginning of vapour condensation $r_{b2}(0)$ (approximated as the $r_b(t_1)$),

$$\frac{1}{3} \left[\frac{r_{b2}(t)}{r_{b2}(0)} \right]^2 + \frac{2}{3} \frac{r_{b2}(0)}{r_{b2}(t)} = 1 + \frac{t}{t_{cond}}, \quad t_{cond} = \frac{\pi [r_{b2}(0)]^2}{4Ja^2 a_l} \quad (5)$$

where t_{cond} is the condensation characteristic time and Ja is the Jakob number, $Ja = \frac{\rho_l c_l (T_{bs} - T_{sat})}{\rho_v L_v} (T_{bs} - T_{sat})$ the bubble surface temperature, T_{sat} the saturated temperature, $L_v = 1.02 \times 10^7 J \cdot Kg^{-1}$ the latent heat of evaporation, $\rho_v = 1850 Kg \cdot m^3$ the vapour density).

In stage (2), the enriched hydrogen in melt pool may diffuse into the bubble driven by the concentration difference. The bubble size r_{b3} induced by hydrogen diffusion may be estimated by the characteristic length of hydrogen diffusion limited growth l_D ³³ as, $r_{b3} = l_D/2$, $l_D = \sqrt{D_h t}$, where D_h is the mass diffusivity of hydrogen in liquidus aluminium and may be approximated as an average of $D_h(T_l) = 1.0943 \times 10^{-7} m^2 \cdot s^{-1}$ and $D_h(T_v) = 1.1302 \times 10^{-5} m^2 \cdot s^{-1}$ based on reference⁶⁹. Accordingly, the bubble radius in stage (2) that is controlled by vapour condensation and hydrogen diffusion may be described as, $\frac{4}{3} \pi r_b^3 = \frac{4}{3} \pi r_{b2}^3 + \frac{4}{3} \pi r_{b3}^3$, based on mass balance (assuming that the contents, temperature and pressure within the bubble are homogeneous). Therefore, the instantaneous bubble radius in stages 1 and 2 can be approximated as,

$$r_b(t) = \begin{cases} r_{b0} + \Delta r_{b1}(t), & t \leq t_1 \\ \sqrt[3]{[r_{b2}(t - t_1)]^3 + [r_{b3}(t - t_1)]^3}, & t > t_1 \end{cases} \quad (6)$$

All the parameters used in the model were listed in Supp. Table S. 2 (thermal-physical properties) and Supp. Table S. 3 (processing parameters), which were used to plot the bubble diameter graph in Fig.4b, c. Note that we used the liquidus aluminium temperature $T_l = 933.5 K$ to approximate the bubble saturation temperature T_{sat} in melt pool, while for the bubble surface temperature T_{bs} , which is subcooled by uncertainty temperature in the melt pool after the bubble being pinched off from keyhole, we approximated its magnitude by fitting between observed data and the modelled results. For a spherical vapour bubble,

the total condensation time is around $1 \sim 3 t_{cond}$ ³⁰. Here, we used the average $2t_{cond}$ and modelled the bubble dynamics within $2t_{cond}$.

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Author contributions

P.D.L. and Y.H. conceived the project. Y.H. wrote the manuscript with significant contributions from P.D.L, C.L.A.L., T.G.F., and S.J.C., performed the characterization, image processing (with help from C.L.A.L. and J.T.), performed data analysis and modelling. P.D.L., C.L.A.L., S.J.C., S.M. and K.F. led and performed the experiments. T.G.F. calculated the peak-to-peak period and frequency for keyhole width/depth. S.J.C programmed the Kalman filter tracking.

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