Enhanced singular jet formation in oil-coated bubble bursting

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Bubble-bursting aerosols have a key role in mass and momentum transfer across interfaces. Previous studies report that the bursting of a millimetre-sized bare bubble at an aqueous surface produces jet drops with a typical size on the order of 100 μm. Here we show that jet drops can be as small as a few micrometres when the bursting bubble is coated by a thin oil layer. The faster and smaller jet drops result from the singular dynamics of the oil-coated cavity collapse. The air–oil–water compound interface offers a distinct damping mechanism to smooth out the precursor capillary waves during cavity collapse, leading to a more efficient focusing of the dominant wave and thus allowing singular jets over a much wider parameter space than that of a bare bubble. We develop a theoretical explanation for the parameter limits of the singular jet regime by considering the interplay between inertia, surface tension and viscous effects. Contaminated bubbles are widely observed, therefore previously unrecognized fast and small contaminant-laden jet drops may contribute to the aerosolization and airborne transmission of bulk substances.

Bubbles present in liquids are commonplace in a wide spectrum of natural and industrial processes1–8. In cases where bubbles rise to the surface of the liquid, they burst and generate numerous droplets, including jet drops and film drops8,9. The jet drops are formed by the fragmentation of an upward jet induced by cavity collapse, while the film drops are produced from the disintegration of the bubble cap. These drops play an important role in many transport processes across the air–liquid interface4,7,10–12. For example, drops from bursting bubbles are considered to be the main source of sea spray aerosols10 and impact air pollution11,12, global climate4,13,14 and even the transmission of infectious diseases15–17. Although most previous studies focused on clean bubbles, in practice bubbles with a compound interface are more ubiquitous. Such bubbles could be formed as bubbles in natural water bodies scavenge surface-active organic materials while they rise18. Other examples include gas bubbles released from natural seeps19, froth flotation20 and material processing using coated bubbles21,22. Within this context, it remains unclear how a compound interface, for example one that is formed by an insoluble coating on the surface of a contaminated bubble, mediates the bubble-bursting dynamics and the related mass and momentum transport.

Previous works claim that a millimetre-sized bare bubble bursting in water produces jet drops of order O(100 μm) (with a typical ejection velocity of O(1 m s−1))25,26, which are unlikely to contribute to bubble-driven aerosols because of the short floating duration6. However, we show that the bursting of a millimetre-sized bubble contaminated by an oil coating (Fig. 1a–c and Extended Data Fig. 1) on clean water can generate micrometre-sized jet drops with an ejection velocity as large as O(10 m s−1). Figure 1c shows that such an extremely thin and fast jet emerges above the water surface after bubble cap rupture, and then breaks up into multiple jet drops. We further confirmed that the jet drops consisted of oil only when the oil volume fraction (defined as the ratio between the oil and bubble volumes, see Methods) ψo ≥ 0.5% by checking the composition of the jet drops using a test strip for water detection (see Methods and Extended Data Fig. 2). Such fast and thin jets were not observed for a bare bubble bursting in a pure water or oil phase (Fig. 1d and Extended Data Fig. 3). Similar to the film drops, micrometre-sized oily jet drops are noteworthy because their slow

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settling velocities allowed them to persist long and travel far. Thus, they unavoidably affect the chemical compositions of the sea spray and the airborne transmission of bulk substances such as contaminants and viruses. Our findings suggest that the role of the jet drops in bubble-bursting aerosols should be carefully revisited for compound bubbles.

The fast, thin jets observed here are often referred to as singular jets, which are found to result from finite-time self-similar dynamics of the cavity collapse. Side-view high-speed observations show how oil-coated cavity collapse leads to the formation of a singular jet (Fig. 1e) distinct from bare bubble bursting (Fig. 1f). The compound bubble is initially fully engulfed by silicone oil. After the bubble cap ruptures, a train of capillary waves is excited and travels downwards along the air–oil–water interface. As the oil is swept towards the nadir of the bubble cavity, the accumulation of oil and the ejected jet morphology were measured when the jet crossed the undisturbed air–water interface (inset of Fig. 2a). For oil-coated bubbles with \( \mu_o = 1.8 - 9.3 \) mPa s, singular jets occurred when \( \psi_o > 2\% \), as \( \psi_o \) increased rapidly with a sharply decreasing \( r \). Meanwhile, for \( \mu_o = 0.9 \) or 19 mPa s, no jet singularity was observed for any value of \( \psi_o \). These singular jets are characterized by narrow and fast jets with high peak jet velocities that are sensitive to the initial bubble rupture process. In addition, we note that we experimentally and theoretically found that \( h_{\text{max}}/R \propto \psi_o^{2/3} \), where \( h_{\text{max}} \) is the maximum oil layer thickness at the bottom pole of the bubble (Fig. 1a and Supplementary Section 3).

Inspired by previous studies, we define singular jets as those with a dimensionless tip radius \( r/R \leq 0.025 \), which corresponds to an effective surface tension, \( \gamma = \gamma_o + \gamma_{wo} \), where the sum of the oil–air and oil–water interface tensions and \( \rho_o \) is the density of the aqueous phase; \( \psi_o \) and \( r \) were measured when the jet crossed the undisturbed air–water interface (inset of Fig. 2a). For oil-coated bubbles with \( \mu_o = 1.8 - 9.3 \) mPa s, singular jets occurred when \( \psi_o > 2\% \), as \( \psi_o \) increased rapidly with a sharply decreasing \( r \). Meanwhile, for \( \mu_o = 0.9 \) or 19 mPa s, no jet singularity was observed for any value of \( \psi_o \) (Supplementary Fig. 1). The evolution of the jet morphology with \( \mu_o \) and \( \psi_o \) is shown in Fig. 2e–c.

To gain a quantitative understanding of how the oil coating facilitates the formation of jet singularities for the case of coated bubble bursting, numerical simulations were performed using the open-source software Basilisk (Methods). As in previous studies, the initial condition we used was given by the static shape of an oil-coated bubble obtained by solving the Young–Laplace equation (Methods and Extended Data Fig. 5). The simulations captured the evolution of the bubble cavity, the accumulation of oil and the ejected jet morphology reasonably well (Extended Data Fig. 6). These results thus provide an answer to the question of how the coating facilitates the formation of jet singularities for the case of coated bubble bursting.
We propose that the occurrence of the singular jet requires that all of the precursor waves at the air–oil interface, including the SW, are sufficiently damped for the DW to maintain self-similar focusing before the viscous effect directly limits the jet velocity after jet formation. Furthermore, the SW can serve as an indicator of this transition because it has the longest wavelength, corresponding to the smallest damping rate in all precursor waves. The strength of the SW was measured from the simulations using the dimensionless maximum principal curvature $\kappa = R/r_e$, ahead of the DW (inset in Fig. 3a) where $r_e$ is the minimum radius of curvature. A smaller $\kappa$ indicates a weaker capillary wave. We observed that $\kappa$ decreased as $\mu_o$ or $\psi_o$ increased, resulting from stronger viscous dissipation effects (Fig. 3d,e). Unlike the case of bare bubble bursting, a further decrease in $\kappa$ was observed for compound bubble bursting when the local oil thickness $h_o/R$ (see the definition of $h_o$ in Fig. 3a) exceeded a value of approximately 0.04 (Fig. 3d,e). The enhanced damping effects can be interpreted as the SW fully propagating into the air–oil interface and thus experiencing a more viscous oil layer. Here we considered $h_o/R$, as the wavelength of the capillary waves generated by bubble cavity collapse was found to scale with $R$ (refs. 12,24). For all singular cases, $\kappa$ eventually reached a value of ~10–30, consistent with the previous observation of the SW for singular jet formation from bare bubble bursting.

To quantitatively describe the prerequisites for singular jets, we further evaluated the viscous damping rate of the DW and SW during cavity collapse. The viscous damping rate for the amplitude deviations of at least three measurements. c. Regime map of jet singularity with $\psi_o$ and $\mu_o$. The red and blue dashed lines correspond to the experimental cases in d and e, respectively. d, e. Experimentally observed jet morphology for different $\mu_o$ with $\psi_o = 2.3 \pm 0.6$% (d) and different $\psi_o$ with $\mu_o = 4.6$ mPa s (e). Scale bars, 1 mm.
of a capillary wave with wavelength $\lambda$ at a free liquid surface can be estimated as $^{34}$:

$$T^{-1} = \frac{8\pi^2 \mu}{\rho \lambda^2}$$  \quad (1)

For bare bubble bursting, the DW and SW have wavelengths of $\lambda_s = R$ and $\lambda_c = 0.25R$, respectively. Therefore, the damping rates of the SW ($T^{-1}_{\lambda_s}$) and DW ($T^{-1}_{\lambda_c}$) during bare bubble bursting are correlated by a constant ratio $\zeta = T^{-1}_{\lambda_c}/T^{-1}_{\lambda_s} = (\lambda_c/\lambda_s)^2 \approx 16$. However, for oil-coated bubble bursting, we show that $\zeta$ can be substantially increased due to the compound interface. To gain insight into the capillary wave dynamics in the complex, non-uniform oil layer around the collapsing cavity, we considered a simplified set-up consisting of a capillary wave propagating at a free aqueous surface covered by a uniform oil layer of thickness $h$. We then developed a wave damping model based on the linear capillary wave theory to calculate $\zeta$ in this configuration (Fig. 4a and Methods). As shown in Fig. 4b, $\zeta$ approaches $(\lambda_c/\lambda_s)^2$ when $h$ approaches 0 or $\infty$, with a maximum located at $h = O(\lambda_c)$ (Supplementary Section 4). This non-monotonic behaviour shows that a more viscous oil layer coating the bubble cavity with $h = O(\lambda_c)$, corresponding to the current experiments, may greatly increase the ratio of the damping rates between the SW and DW relative to that at a clean interface. Therefore, the compound interface favours the production of a singular jet by smoothing out the shorter-wavelength perturbations.

In addition, in the case of oil-coated bubble bursting, the capillary waves encounter an oil layer with non-uniform thickness, which leads to a capillary wave separation, thus further increasing the damping rate ratio between the SW and DW. This can be seen by considering the wave speed and wavelength of the SW during the oil-coated cavity collapse. The wave speed is set during the initial film rupture at the top air–oil–water interface, given by $U = 5U_{crit}$, as confirmed by the experiments and simulations (Extended Data Fig. 7c). As the SW propagates, it encounters an oil layer of increasing thickness and splits between the air–oil and oil–water interfaces while maintaining a nearly constant wave speed. As the oil layer thickness increases, the SW begins to experience a different bulk liquid with a different surface tension, thus further increasing the damping rates between the SW and DW relative to that at a clean interface.
The damping rate of the SW and the bubble collapse time for oil-coated bubbles is given by the relationships $\psi = (2)$ and $h = (\text{wavelength})$, respectively. The inset shows the model configuration with a capillary wave with $h$ and $\mu$. The proposed Oh model calculation for SW damping at different $\psi$ and $h/\lambda_c$ vary inversely with the inertia–capillary timescale (that is, $\zeta = 2\pi \sqrt{\rho R^3/\gamma}$). Comparison between successive cavity profiles rescaled (left) and unrescaled (right) with $t_0 = t_0^{1/3}$, where $\mu_w = 1.8$ mPa s and $\gamma = 4.2\%$. Here $r^* = r (\gamma \rho_0 / \rho_0)^{-1/3} (t_0 - t)^{-2/3}$ and $z^* = (z - z_0) (\gamma \rho_0 / \rho_0)^{-1/3} (t_0 - t)^{-2/3}$, where $t_0$ represents the moment when the jet forms and $z_0$ represents the bottom location of the entrapped bubble at $r_0$. The inset shows that $r_0$ versus time before the jet follows a power law of 2/3 (dotted line). Time-lapse images of drop ejection (top) from collective bursting (bottom) of oil-coated bubbles in pure water ($\mu_w = 4.6$ mPa s, left), bare bubbles in pure water (middle) and bare bubbles in 0.8 mM sodium dodecyl sulfate solution (right, to mimic the natural environment enriched with surface-active compounds). The bubbles are generated with a frequency of $2 \text{ s}^{-1}$. Scale bar, 10 mm.

**Fig. 4** Regime map of singular jets and jet drop generation by collective oil-coated bubble bursting. **a,** Schematics of precursor waves during bubble bursting. The inset shows the model configuration with a capillary wave with wavelength $\lambda$ at a compound interface where the top liquid has a uniform thickness of $h$. **b,** Variation in $\zeta$ with $h/\lambda_c$ calculated by the proposed model. Here, $\mu_w/\mu_o = 5$ and $\lambda_c = R = 2$ mm. The grey shaded area represents the range of $h$ observed in the oil-coated bubble experiments and simulations. **c,** Regime map of singular jets in both experiments (empty symbols) and simulations (filled symbols). The bounding criteria are (I) $\text{Oh}_r = 11$ (see Methods) and (II) $\text{Oh}_o = 0.06$. The shading indicates the singular (S, blue) and non-singular (NS, orange) domains. **d,** Comparison between successive cavity profiles unrescaled (left) and rescaled (right) with $(t_0 - t)^{1/3}$, where $\mu_w = 1.8$ mPa s and $\gamma = 4.2\%$. Here $r^* = r (\gamma \rho_0 / \rho_0)^{-1/3} (t_0 - t)^{-2/3}$ and $z^* = (z - z_0) (\gamma \rho_0 / \rho_0)^{-1/3} (t_0 - t)^{-2/3}$, where $t_0$ represents the moment when the jet forms and $z_0$ represents the bottom location of the entrapped bubble at $r_0$. The inset shows that $r_0$ versus time before the jet follows a power law of 2/3 (dotted line). **e,** Time-lapse images of drop ejection (top) from collective bursting (bottom) of oil-coated bubbles in pure water ($\mu_w = 4.6$ mPa s, left), bare bubbles in pure water (middle) and bare bubbles in 0.8 mM sodium dodecyl sulfate solution (right, to mimic the natural environment enriched with surface-active compounds). The bubbles are generated with a frequency of $2 \text{ s}^{-1}$. Scale bar, 10 mm.

The model calculation for SW damping at different $\psi$ and $h/\lambda_c$ vary inversely with the inertia–capillary timescale (that is, $\zeta = 2\pi \sqrt{\rho R^3/\gamma}$). Comparison between successive cavity profiles rescaled (left) and unrescaled (right) with $(t_0 - t)^{1/3}$, where $\mu_w = 1.8$ mPa s and $\gamma = 4.2\%$. Here $r^* = r (\gamma \rho_0 / \rho_0)^{-1/3} (t_0 - t)^{-2/3}$ and $z^* = (z - z_0) (\gamma \rho_0 / \rho_0)^{-1/3} (t_0 - t)^{-2/3}$, where $t_0$ represents the moment when the jet forms and $z_0$ represents the bottom location of the entrapped bubble at $r_0$. The inset shows that $r_0$ versus time before the jet follows a power law of 2/3 (dotted line). Time-lapse images of drop ejection (top) from collective bursting (bottom) of oil-coated bubbles in pure water ($\mu_w = 4.6$ mPa s, left), bare bubbles in pure water (middle) and bare bubbles in 0.8 mM sodium dodecyl sulfate solution (right, to mimic the natural environment enriched with surface-active compounds). The bubbles are generated with a frequency of $2 \text{ s}^{-1}$. Scale bar, 10 mm.

On the basis of our modelling of capillary wave damping, we rationalized the bounding criteria for singular jets from compound bubble bursting. For bare bubble bursting, Oh can be interpreted as $\text{Oh} \propto T_{\lambda_c}^{-1} \zeta^{-1}$, the ratio between the damping rate of the SW and the inverse of the inertia–capillary timescale (that is, $\zeta^{-1} = (\rho R^3/\gamma)^{-1/3}$). However, for compound bubble bursting, we obtained $T_{\lambda_c}^{-1}$ with our model calculation for SW damping at different $\psi$ and $\text{Oh}_o$, and we propose a revised Oh:

$$\text{Oh}_o = \frac{T_{\lambda_c}^{-1}}{T_{bc}^{-1}}$$

where $T_{bc} = 0.3 \sqrt{\rho R^3/\gamma}$ is the bubble collapse time for oil-coated bubbles obtained from our experiments and simulations. We found that the isoline of $\text{Oh}_o = 11$ aligned well with the lower and left boundaries of the singular jet regime (Fig. 4c), which is quantitatively analogous with the lower critical $\text{Oh} = 0.03$ for singular jets from bare bubble bursting in a single liquid (Methods and Supplementary Section 5). The SW damping is therefore responsible for setting boundary (I) ($\psi > 1\%$ and $\text{Oh}_o > 0.01$) in Fig. 4c, which is captured by our proposed $\text{Oh}_o$. In addition, the numerical results for singular jetting confirm that the minimum cavity radius follows the inertia–capillary self-similarity behaviour with the power law $r_c \propto (t_0 - t)^{1/3}$ (Fig. 4d).

Furthermore, with a further increase in $\text{Oh}_o$, the viscous stresses continuously dampen the DW, limiting the jet velocity and enlarging the top jet drop due to the delay of jet break-up. This excess viscous damping results in a maximum $\text{Oh}_o$ for which singular jetting can occur. This transition is shown as boundary (II) in Fig. 4c and corresponds to $\text{Oh}_o = 0.06$, consistent with the transitional value of $\text{Oh} = 0.03$ for singular jetting in bare bubble bursting when the viscous stresses become strong enough to directly suppress cavity cusp formation. In addition, when...
the bulk viscosity $\mu_w$ varies from 1 to 22.5 mPa s, corresponding to $Oh_{\nu_w} = \sqrt[3]{\frac{\mu_w R}{\rho_0 f}} \cdot (10^{-10})$, singular jetting occurs when $\psi_1 < 1$ (Extended Data Fig. 8), whereas bare bubble bursting only produces singular jets within a narrow range of $Oh < 0.02–0.05$ [19] for the bulk liquid. These results show that the compound interface with the oil coating could facilitate the inertia–capillary self-similarity, expanding the range of singular jetting in bubble bursting and decreasing the jetted drop sizes.

More generally, our study on oil-coated bubble bursting demonstrates the role of the compound interface on the bubble-driven aerosol flux. In particular, owing to the wider parameter space for singular jetting, collective oil-coated bubble bursting tends to generate jet drops with smaller sizes, greater overall numbers of drops and higher droplet ejection heights than bare bubble bursting at either clean or surfactant-laden aqueous surfaces, as shown in Fig. 4e. Here a sodium dodecyl sulfate solution was used to mimic a natural water body enriched with surfactant-active compounds. The droplet size is one key parameter in predicting its residence time and transport, as small droplets are more easily lifted by turbulent eddies [18]. These contaminant-laden drops smaller than 10 μm in diameter could pose a higher risk of pollutant spread or infection as they can penetrate farther into the respiratory tract than larger drops [19][25]. The oil-coated bubbles in our experiments could typify the ubiquitous contaminated or compound bubbles in the oceans, and bubble-bursting jet drop particles have been found to contain different compositions with stronger ice nucleating abilities than film drop particles [19]. Our discovery may therefore improve chemical transport modelling related to bubble-driven fluxes in the context of sea spray aerosols. In industry, these small drops resulting from the singular jets produced by compound bubble bursting may have detrimental impacts on workers’ health, such as the generation of acidic mists in electrolysis [19][26] and bioaerosols from wastewater treatment plants [21][26]. Our work may suggest additional guidelines for personal protective equipment and management controls on air and water quality near these facilities [19]. Bubble bursting is also considered to be the major cause of cell damage in bioreactors via the hydrodynamic stresses produced by cavity collapse and jet break-up [19]. The thin and fast singular jet regime from compound bubbles may sharply increase these stresses, and thus require more careful control of aeration and agitation. In summary, these results for the production of singular jets from oil-coated bubble bursting offer insights into the dynamic processes of complex fluids, with potential environmental consequences and industrial applications.

**Online content**

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-023-01958-z.

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Methods

Materials
Deionized water (with a resistivity of 18.2 MΩ cm) was obtained from a laboratory water purification system (Smart2Pure 3 UV/UF, Thermo Fisher Scientific). Octamethyltrisiloxane (referred to as silicone oil, with a kinematic viscosity \( v = 1 \times 10^{-4} \text{ m}^2 \text{s}^{-1} \)), dodecamethylpentasiloxane (silicone oil with \( v = 2 \times 10^{-8} \text{ m}^2 \text{s}^{-1} \)), other silicone oils (\( v = 5 \times 10^{-5}, 1 \times 10^{-4}, 2 \times 10^{-4} \text{ m}^2 \text{s}^{-1} \)) and sodium dodecyl sulfate (BioXtra, \( \geq 99.0\% \)) were purchased from Sigma-Aldrich and used as received. Glycerin was purchased from Fisher Chemical. The surface tensions of oil \( \gamma_{\text{oa}} \) (or surface tensions of water or aqueous solutions \( \gamma_{\text{aw}} \)) and the interfacial tensions between silicone oils and water \( \gamma_{\text{ow}} \) were measured using the pendant drop method, and the densities \( \rho \) and dynamic viscosities \( \mu \) are listed in Extended Data Table 1.

Experimental set-up

The experimental apparatus is shown in Extended Data Fig. 1. A square transparent acrylic container of \( 20 \times 20 \times 25 \text{ mm}^3 \) was fabricated to hold the liquids, and we measured the contact angle of water on the acrylic as \( 86 \pm 6^\circ \). We used a custom-designed coaxial orifice system \(^{51,52} \) to produce oil-coated bubbles. The system consisted of a small stainless inner needle with an inner diameter of 0.41 mm, plugged into a stainless outer needle with an outer diameter of 3.43 mm. A sessile oil droplet was first formed by injecting oil between the two needles, then air was injected through the inner needle to form the oil-coated bubble. The equilibrium radius of the compound bubble (gas + oil) in our experiments was determined to be \( R = 2 \text{ mm} \).

Two high-speed cameras (FASTCAM Mini AX200, Photron) were used to synchronously record the top and bottom views of the oil-coated bubble bursting at a free liquid surface, separately illuminated by two LED panels. We carefully maintained a slightly convex meniscus at the air–water interface over the container edge by filling up the container. This slightly convex meniscus prevented the bubble from drifting away from the focal plane, and also allowed better imaging of the jet.\(^{51} \) In addition, by tilting the cameras at an angle of \( -5^\circ \), the influence of the meniscus on the visualization could also be avoided. We used a frame rate of 6,400–20,000 frames per second and magnification of \( x \approx 4 \). We also used an advanced high-speed camera (FASTCAM SA-Z, Photron) with a frame rate of 50,000 frames per second and a magnification of \( x \approx 12 \). To obtain high-resolution images, the obtained images were post-processed with Fiji (Imagej 1.52p)\(^{48} \) and MATLAB 2020a (Mathworks). The volume of the oil in the oil-coated bubble \( V_o \) was estimated by measuring the oil volume at the bubble bottom in the high-speed video before jet formation, and then the oil fraction was calculated as \( \phi_o = 3V_o/(4\pi R^3) \).

For the collective bubble bursting (Fig. 4e), bare gas bubbles were generated with a 3.43-mm-diameter needle. The equilibrium gas bubble radius was determined to be \( 2.3 \pm 0.2 \text{ mm} \), similar to the oil-coated bubble radius. In each experiment, we took a high-speed video with a duration of \( 11 \text{s} \) at a frame rate of 125 frames per second. All top-view images were overlapped to produce the upper row of Fig. 4e.

To provide more information on the composition of the jet drop, a cobalt chloride test strip for water detection (PGA01V100, Bartovia) was used to collect the jet drops by bubble bursting to test for the presence of water. If the drop contact location turned pink, the jet drop contained water.\(^{50} \) We note that we performed control experiments with micropipette tips to manipulate the deposited drop size, and found that the colour change was observable for a water drop with a radius as small as 15 \( \mu \text{m} \). In our bubble-bursting experiments with 4.6 mPa s oil, when \( \phi_o = 0.5\% \), the jet drop radius was larger than 100 \( \mu \text{m} \). There were no colour changes of the test strips. Thus, the jet drops should only contain oil for \( \phi_o \geq 0.5\% \) as our control experiments showed that water could be detected in drops with a radius of 15 \( \mu \text{m} \). This critical \( \phi_o \) was further confirmed by our numerical simulation with 4.6 mPa s oil (Extended Data Fig. 2), in which the oil volume composition of the top jet drop was \( -10\% -40\% \) when \( \phi_o < 0.5\% \) and \( 100\% \) when \( \phi_o \geq 0.5\% \).

Numerical simulations

Numerical simulations were performed using the open-source partial differential equation solver Basilisk\(^{56,59} \). The Basilisk solver is especially well suited to performing adaptive mesh refinement, which is critical for resolving such multiphase flow problems with jetting\(^{12} \), droplet break-up\(^{9} \), drop impact and splash\(^{60,61} \) and thin films.\(^{56} \) In particular, axisymmetric simulations were performed using the three-phase solver developed by Sanjay et al.\(^{63} \).

For the simulations, we set the initial conditions as an oil-coated bubble resting at an air–liquid interface with a hole to connect the bubble interior to the gas phase above the interface, which represents a symmetrically rupturing bubble cap. We developed a model to calculate the initial shape of the static oil-coated bubble (see ‘Initial static bubble shape’ and Supplementary Section 2) that precisely reproduced the experimental static bubble shape as shown in Extended Fig. 5. In each case, the oil-coated bubble was initialized in a 15 mm square domain where \( r \) ranged from 0 to 15 mm and \( z \) ranged from \(-7.5 \) to 7.5 mm. The \( z \) origin of the bubbles was shifted to give sufficient room to resolve the jetting drops and so the water at the bottom of the domain was relatively undisturbed. We used a minimum refinement level of 5, corresponding to a maximum simulation cell size of \( \approx 0.469 \) mm and a maximum refinement level of 13, corresponding to a minimum simulation cell size of \( \approx 0.83 \mu \text{m} \). The initial condition was always resolved up to the maximum refinement level, and then the adaptive mesh refinement took over, always resolving any fluid interfaces to the maximum level. Specifically, the adapt function was used to control the adaptive meshing at each time step with tolerance values on the interface volume fraction fields, the velocity components and the curvatures of the air–oil and oil–water interfaces of \( \times 10^{-3} \) for each tolerance. The imposed non-default boundary conditions in the simulations were a zero normal gradient condition on the velocity and a fixed zero Dirichlet boundary condition at the top boundary. The solver automatically controlled the time step to guarantee stability based on the surface tensions, velocities and adaptive meshing, and output data files were written every 0.1 or 0.01 ms. A pre-wetting oil film with a layer of exactly one cell thickness was assumed at the air–water interface\(^{43,44} \).

For the configuration with an oil-coated bubble bursting in water, that was thermodynamically favourable for oil to spread on water (that is, the spreading coefficient \( S = \gamma_{\text{ow}} - \gamma_{\text{oa}} - \gamma_{\text{aw}} \geq 0 \) (refs. \(^{62,63,64} \))), as in our current simulations with silicone oil and water. A typical runtime of \( \approx 500 \) h of CPU time was used for each of these cases, and we ran each simulation in parallel on 32 processors.

The numerical simulations were validated by comparing with experiments with bare bubble bursting at pure liquid surfaces regarding the jet tip radius and velocity. For the oil-coated bubble, our simulation showed good agreement with experimental results with respect to the time for cavity collapse and overall shape of the interfaces (Extended Data Fig. 6). We also performed simulations with different refinement levels as convergence tests to confirm that our results were independent of the mesh refinement level. With the current refinement level of 13, the velocity and radius of the non-singular jets and the width of the collapsing cavity shape were converged (the relative errors remained within 1% when the maximum refinement level was increased from 12 to 13). However, the singular jet velocity and radius did not converge as the refinement level was increased. Therefore, we considered the convergence tests as distinguishing between regimes that illustrate singular jet formation and the regimes that do not. To that end, we performed simulations with maximum refinement levels of 11, 12 and 13 and found that they all predicted the same singular jet regimes (see detailed discussion in Supplementary Section 6). In that sense, and for the purpose of the current work, we consider these
results converged, and we opted for the higher refinement level for the best accuracy possible. We note that for the bare bubble bursting, a maximum level of 13 in simulations with Basilisk has been adopted by previous work on bubble-bursting jets. \textsuperscript{40,44} In addition, our simulations assumed an axisymmetric flow as most previous simulation work has done,\textsuperscript{43,44} but bubble bursting in reality might show asymmetry from film rupture to the final jet formation and break-up.\textsuperscript{26} Furthermore, it has been suggested that predictions of the singular jet velocity and radius from simulations are difficult to compare with experiments due to the convergence issues.\textsuperscript{25} All of the above factors may contribute to the discrepancies between the experiments and simulations, particularly regarding the singular jet tip velocity and radius.

Within the scope of the current work, we observed strong agreement between the numerically and experimentally determined parameter regimes for singular jet formation (Fig. 4c). In addition, regarding the propagation of the SW contributing to the jet formation, we also obtained good consistency, as shown in Extended Data Fig. 7. Therefore, we believe that our simulations provide reliable and insightful information with which to understand the jet formation in oil-coated bubble bursting.

**Initial static bubble shape**

The initial static shape of an oil-coated bubble resting at the water surface was calculated by numerical solutions of the Young–Laplace equation. The assumed static bubble shape was separated into several interface portions and solved iteratively with matching boundary conditions, following a similar approach to that used in previous work\textsuperscript{26} (see Supplementary Section 2).

**Capillary wave damping model**

To rationalize the damping of the capillary waves during the oil-coated cavity collapse, we present a simplified model to describe the effect of the oil coating on cavity collapse and jet formation. As shown in Fig. 4a, a layer of oil with a uniform thickness \( h \) at rest was deposited on a deep bath of water. Here, we neglected the bubble cavity curvature and film thickness variation considering the fact that \( h_{\text{sw}}/R \approx 1 \) in the experiments. In addition, the gravitational effects were negligible given that the typical capillary wavelength in bubble bursting is smaller than the capillary length \( \sqrt{\rho_{\text{oa}}/\gamma} \). We considered a periodic traveling capillary wave with the longwave approximation, which has been widely used to model the waves excited by bubble cavity collapse.\textsuperscript{43,44} In addition, we assumed that nonlinear effects were negligible to use the linearized Navier–Stokes equations. This can be justified because the boundary layer that develops around the wave crest has a thickness \( \delta \approx \sqrt{2\rho_{\text{oa}}/\gamma} \), where the angular frequency \( \omega \) can be estimated as \( \sqrt{(\gamma_{\text{oa}}/\rho_{\text{oa}})(2\pi/\lambda)^{3}} \) (refs. \textsuperscript{32,64}). In all our experiments, \( \delta/\lambda < 0.2 \). Following the linear capillary wave theory,\textsuperscript{44,70,71} we derived the dispersion relation using the linearized Navier–Stokes equation (Supplementary Section 4). By numerically solving the dispersion relation, we obtained the wave damping rate, which was further validated with previous theoretical results\textsuperscript{44} in the limit of \( h \to 0 \) (Supplementary Section 4). We used our derivation to estimate the damping rate of the SW and DW for the oil-coated cavity collapse and calculate \( \text{Oh}_{s} \).

Notably, our linear wave damping model has limitations with respect to short capillary waves where the longwave approximation might not hold, or for highly viscous liquids where \( \delta \approx \lambda \). In the latter case, the viscous damping in the rotational flow within the boundary layer needs consideration, and thus nonlinear effects cannot be neglected.\textsuperscript{44,45}

**Capillary wave separation**

Here we focus on further characterization of the capillary wave propagation during cavity collapse as these waves directly dictate the formation of the jet. Extended Data Fig. 7a,b shows the propagation of capillary waves in the case of a compound or bare bubble bursting and the SW is highlighted. While the wave speed remains almost the same, the wavelengths and amplitudes of the SW in the compound bubble case are smaller. The capillary wave speed \( U \) and \( \lambda \) are associated with the dispersion relationship\textsuperscript{44} as:

\[
\lambda = \frac{2\gamma}{\rho U^2}.
\]

which has been applied to analyse the capillary wave propagation resulting from bubble cavity collapse with a small \( \text{Oh} \).\textsuperscript{44,45} The variation of \( \theta \) through time has been used to characterize the wave propagation speed as \( U = \text{Oh}/\theta \) (ref. \textsuperscript{32}).

For bare bubble bursting in pure liquids, \( U/\text{Oh} = 5 \), where the characteristic capillary velocity for a clean bubble is \( \text{Oh} = \sqrt{\gamma_{\text{oa}}(\rho_{\text{oa}}R)} \) (refs. \textsuperscript{25,32,35}). Thus, the wavelength of the SW for bare bubble bursting, \( \lambda_{\text{bp}} \), is obtained as:

\[
\lambda_{\text{bp}} = \frac{2\pi}{25}.
\]

We note that this prediction is consistent with the previous work\textsuperscript{35,42}. Regarding oil-coated bubbles with a compound interface, the capillary waves split into the air–oil and oil–water interfaces when the oil layer becomes thick. However, during the whole cavity collapse, \( U/\text{Oh} \) was also found to be 5.7 ± 0.7 in both our experiments and simulations, as shown in Extended Data Fig. 7c. When the SW has fully entered the air–oil interface after the wave separation, the oil density \( \rho_{o} \) and surface tension \( \gamma_{o} \) are to be considered in equation (3), so we obtained:

\[
\frac{\lambda_{\text{sp}}}{R} \approx \frac{\rho_{o} Y_{oa}}{\rho_{c} Y_{c}} \approx \frac{\rho_{o} Y_{oa}}{\rho_{c} Y_{c}} \frac{\lambda_{\text{bp}}}{R}.
\]

With simulations, we showed that the dimensionless SW wavelength of compound bubble bursting at \( \theta = \pi/6 \) decreased notably as \( \lambda_{\text{sp}}/R = 0.11 \) after capillary wave separation for large \( \psi_{c} \) compared with that for a bare bubble, which is \( \lambda_{s}/R = 0.28 \) (Extended Data Fig. 7d). The measurements agreed well with our prediction in equations (4) and (5). Our results confirmed that the compound interface also contributed to the decrease in the SW wavelength, and thus increased the corresponding damping rate.

**Oh, for singular jets**

It has been shown that the decrease in the capillary wave amplitude during cavity collapse of bare bubble bursting could be described by Oh\textsuperscript{42,71}. At a free liquid surface, the amplitude \( \alpha \) of capillary waves falls off exponentially in the form \( \alpha = \alpha_{0} e^{-\text{Oh}^{0.5}} \) with the damping rate calculated by equation (1). In the context of bare bubble bursting in a pure liquid, Krishnan et al.\textsuperscript{16} proposed that in the bubble collapse time \( t_{bc} = 0.3\psi_{c} \), where \( \psi_{c} = \sqrt{\gamma_{oa}/\rho_{oa}} \), so the decrease in \( \alpha \) is given by:

\[
\ln\left(\frac{\alpha}{\alpha_{0}}\right) = -\frac{t_{bc}}{T_{bc}} \approx -24\left(\frac{R}{\lambda}\right)^{2}\text{Oh}.
\]

They observed that the capillary waves are progressively damped as \( \text{Oh} \) increases, and the singular jet occurs at a critical Oh due to this damping in bare bubble bursting. Such an observation indicates that when \( \ln\left(\alpha/\alpha_{0}\right) \) increases to a critical value, all the precursor capillary waves are damped and thus the DW is more effectively focused for a self-similar collapse. Therefore, we further considered such a capillary wave damping theory with respect to \( \ln\left(\alpha/\alpha_{0}\right) \) for our oil-coated bubble-bursting jets. We propose a dimensionless number, \( \text{Oh}_{s} \), defined as:

\[
\text{Oh}_{s} = \ln\left(\frac{\alpha}{\alpha_{0}}\right) = \frac{t_{bc}}{T_{bc}}.
\]
which can be interpreted as the damping of precursor capillary waves merging at the bubble base during cavity collapse. $T_c^\alpha$ can be predicted with our proposed model (Supplementary Section S).

As the SW has the longest wavelength among the precursor capillary waves and thus the minimum damping rate, the attenuation of all precursor capillary waves could be described solely by Oh, In both the experiments and simulations, we found that $T_c = 0.3T_c^\alpha$, where $T_c = \sqrt{\rho w R^2 T_c}$, and $T_c$ was calculated with $A/R - 0.1$ using our proposed model for wave damping. The limit of $\text{Oh} = 11$, which indicates the same damping rate of the SW, was found to accurately describe the left and bottom boundaries of the singular regime in the $\alpha$–$\text{Oh}$ regime map for oil-coated bubble bursting. We note that our model does not include any fitting parameters. In addition, considering that $\text{Oh} = -\ln (\alpha/\alpha_1) = 11$ and $A/R = 0.25$ in bare bubble bursting, the lower critical Oh, for singular jetting of bare bubble bursting could be calculated to be $\text{Oh} = 0.03$ using equation (6), which is consistent with the literature values found experimentally and numerically.

Data availability
Source data are provided with this paper and are available via Figsshare at https://figshare.com/articles/dataset/Source_data_English_singular-jet-formation_in_oil-coated_bubble_bursting/2146309. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

Code availability
The codes used for the Basilisk simulation in this study are available at http://basilisk.fr/sandbox/jault/README. The codes for bubble shape calculation are available via GitHub at https://github.com/zyyang-mech/enhanced-singular-jet-formation-in-oil-coated-bubble-bursting.

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Author contributions
B.J. and J.F. conceived the project. B.J. and J.F. designed the experiments. Z.Y. and B.J. conducted the experiments and analysed the results. Z.Y., B.J. and J.F. conducted the theoretical analysis. J.T.A. conducted the simulations with Basilisk. Z.Y. conducted other numerical analyses. J.T.A. and Z.Y. post-processed the simulation results. Z.Y., B.J., J.T.A. and J.F. discussed the results and wrote the paper.

Competing interests
The authors declare no competing interests.

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Extended Data Fig. 1 | Experiment setup for the imaging of oil-coated bubble bursting. a, Schematic drawings of the experiment setup. The oil-coated bubbles are generated from coaxial orifices, and observed with two high-speed cameras simultaneously. b, Zoom-in image of a typical oil-coated bubble at a free surface with $\mu_o = 19$ mPa s and $\psi_o = 6\%$. 
Extended Data Fig. 2 | Variation of oil volume composition in top jet drop from bursting of oil-coated bubbles with different oil volume fractions. The data points denote the oil volume composition of the top jet drop for oil-coated bubble bursting (with 4.6 mPa s oil), obtained from simulations.

Here $\phi_o$ represents oil volume composition in the top jet drop. The dashed line denotes the minimum oil volume fraction to produce an oil-only top jet drop estimated from experiments.
Extended Data Fig. 3 | Bursting of bare bubbles of $R \approx 2$ mm at liquid surfaces with increasing Ohm only produces non-singular jets. A bare bubble with $R = 2.1 \pm 0.3$ mm bursts at the surface of water (Ohm = 0.0026, a), 50 wt% glycerin-water solution (Ohm = 0.015, b), 4.6 mPa s silicone oil (Ohm = 0.025, c), and 70 wt% glycerin-water solution (Ohm = 0.06, d). The scale bar represents 1 mm.
Extended Data Fig. 4 | Dimensionless jet velocity and radius from oil-coated bubble bursting. a, Dimensionless jet velocity $v_j/v_{ce}$ as a function of oil volume fraction $\psi_o$ at different oil viscosities $\mu_o$. For the pure water case, $v_{ce} = v_{cw}$ is used.

b, Dimensionless jet radius $r_j/R$ as a function of $\psi_o$. The hollow markers at the left vertical axes of a, b represent the case for a bare bubble of the same size bursting in pure water.
Extended Data Fig. 5 | Comparison of the numerically calculated static shape of oil-coated bubbles (red dashed curves) with experimental images. The static shapes of the bubbles with $\mu_o = 1.8$ mPa s and $\psi_o = 2.3\%$ (a), 4.3\% (b), and 12.6\% (c) resting at the free surface prior to bursting are well captured by the numerical solutions with the fluid properties listed in Extended Data Table 1. The scale bar represents 1 mm.
Extended Data Fig. 6 | Comparison of the experiment and simulation for oil-coated bubble bursting. Left of each panel shows the experimental high-speed images of an oil-coated bubble bursting. Here $\mu_o = 19$ mPa s, $\psi_o = 4.2\%$, $t = 0$ represents the instant when a hole nucleates in the bubble cap. Right of each panel shows the simulation snapshots of corresponding cavity shape. The white, black, and grey regimes denote air, oil, and water phases, respectively. The scale bar represents 1 mm.
**Extended Data Fig. 7** | Characterization of the SW propagation for oil-coated bubble bursting.  
**a-b.** Capillary wave propagation during the bursting of an oil-coated bubble with $\mu_o = 1.8$ mPa s and $\psi_o = 4.2\%$ (a) and a bare bubble (b). White, black and grey colors represent air, oil, and water phases, respectively. The bubble radius $R = 2$ mm. The scale bar represents 1 mm.  
**c.** Angular wave position $\theta$ as a function of $t^*$ for oil-coated bubble bursting with $R = 2$ mm and $\mu_o = 4.6$ mPa s at different $\psi_o$.  
**d.** Dimensionless SW wavelength $\lambda_s/R$ as a function of $\psi_o$ at $\theta = \pi/6$ for oil-coated bubbles with $\mu_o = 4.6$ mPa s.
Extended Data Fig. 8 | Bubble bursting jet with different bulk liquid viscosities. a-b, Regime map of jet singularity regarding oil fraction $\psi_o$ and bulk liquid viscosity $\mu_w$ (or $Oh_w = \mu_w / (\rho_w R_w a_w)^{1/2}$), with an coating oil viscosity of 1.8 mPa s (a) and 4.6 mPa s (b). c, Experimental snapshots of a singular jet produced by bubble bursting with $\mu_w = 22.5$ mPa s, $\mu_o = 4.6$ mPa s, and $\psi_o = 1.0\%$. The red dashed line marks the bubble cap before rupturing. The scale bar represents 1 mm.
Extended Data Table 1 | Physical properties of the liquids used in the experiments

| Liquids                        | ρ (kg/m³) | μ (mPa s) | γ₁wa (mN/m) | γ₁uw (mN/m) | γ₀uw (mN/m) |
|-------------------------------|-----------|-----------|-------------|-------------|-------------|
| DI water                      | 998       | 1.0       | 71.6 ± 1.0  | N/A         | N/A         |
| 1 cSt silicone oil            | 820       | 0.9       | N/A         | 13.1 ± 0.4  | 27.3 ± 0.3  |
| 2 cSt silicone oil            | 875       | 1.8       | N/A         | 18.6 ± 0.2  | 35.0 ± 0.2  |
| 5 cSt silicone oil            | 913       | 4.6       | N/A         | 18.7 ± 0.3  | 38.1 ± 0.4  |
| 10 cSt silicone oil           | 930       | 9.3       | N/A         | 19.1 ± 0.3  | 40.9 ± 0.5  |
| 20 cSt silicone oil           | 950       | 19.0      | N/A         | 19.4 ± 0.7  | 40.9 ± 0.5  |
| 20 wt% glycerin-water solution| 1047      | 1.8       | 70.9 ± 0.5  | N/A         | N/A         |
| 40 wt% glycerin-water solution| 1099      | 3.7       | 66.9 ± 0.4  | N/A         | N/A         |
| 50 wt% glycerin-water solution| 1126      | 6.0       | 66.4 ± 0.5  | N/A         | N/A         |
| 70 wt% glycerin-water solution| 1181      | 22.5      | 61.4 ± 0.3  | N/A         | N/A         |