We elucidate the evolution of the entrained air in drop impact on a wide range of liquids, using ultrafast X-ray phase-contrast imaging. We elaborate the retraction mechanism of the entrapped air film in terms of liquid viscosity. We found the criterion for deciding if the entrapped air evolves into single or double bubbles, as determined by competition among inertia, capillarity, and viscosity. Low viscosity and low surface tension induce a small daughter droplet encapsulated by a larger air shell bubble, forming an antibubble. We demonstrate a phase diagram for air evolution regarding hydrodynamics.

Drop impact on a liquid surface has great importance in many natural and industrial processes. Raindrops or breaking waves can entrain small air bubbles when they fall onto the sea. This process is a crucial mechanism of gas transport from the atmosphere to the ocean and plays a crucial role in climate change and the ecosystem\(^1\)–\(^4\), and also of great interest in fundamental science and technology. For example, entrapped bubbles can produce underwater noises by their oscillation or enhance nucleate boiling in chemical processes\(^5\)–\(^7\). On solid substrates, air entrapment in drop impact has been actively studied\(^8\)–\(^14\). The air underneath an impacting drop fails to drain and is instead compressed, deforming the bottom surface of the drop. The air layer ruptures thereby, resulting in the entrapment of an air film, eventually evolving into formation of a bubble\(^11\)–\(^14\). On liquid surfaces, however, the evolution of the air entrapment has been largely unexplored because of difficulty in visualizing its microscale (\(< 100 \mu \text{m}\)), rapid (\(< 100 \mu \text{s}\)), and complex dynamics\(^15\)–\(^23\). In particular, there is currently no physical model that exactly predicts the morphology of the entrapped bubble in a wide range of liquid properties and impact conditions.

In this paper, we studied the evolution of the air entrained by drop impact on a variety of liquid pools by using high-speed X-ray phase-contrast imaging that enables us to clearly track the rapid evolution of the interfaces in high temporal (\(< \mu \text{s}\)) and spatial (\(< \mu \text{s}\)) resolutions. The retraction mechanism of an entrapped air film is elaborated in terms of liquid viscosity. The criterion for deciding if the entrapped air evolves into single or double bubbles is rationalized, based on competition among inertia, capillarity, and viscosity. Additionally, it was found that low viscosity and low surface tension induce formation of a small daughter droplet encapsulated by an air shell, resulting in formation of an antibubble. A complete phase diagram for air evolution is demonstrated with respect to hydrodynamic conditions.

**Experiments**

We studied the impact of a liquid drop on a pool of the same liquid with X-ray imaging. To achieve high-intensity light source, white-beam X-ray with a peak irradiance of \(~10^{14} \text{ph/s/mm}^2/0.1\% \text{bw}\) was used\(^24\)–\(^25\). The detector system comprises a fast scintillator crystal (LuAG:Ce, decay time \(~50 \text{ns}\)), a right-angle mirror and the microscope objective (Mitutoyo M Plan APO 10x, \(NA = 0.21\)). The images were captured with a CMOS high-speed camera (Photron Fastcam SA1.1). The imaging speed of the camera was synchronized to the X-ray beam using delay generators, enabling us to capture images with period of 3.68 \(\mu \text{s}\) and exposure time of 472 ns\(^24\)–\(^25\). The drop-impact setup was installed 150 mm distance from the detector to achieve a strong phase-contrast effect. Liquid drops were dispensed from a syringe needle (26 G) connected with a remote-controlled syringe pump, resulting the droplet diameter to be 1.9 ~ 2.6 mm for different liquids. A laser beam was used to sense the drop and trigger the camera and the fast shutter that is installed before the sample stage. The liquid pool for the substrate was prepared...
in a cylinder made by Kapton film with a diameter ~20 mm and a depth ~50 mm, considered as a sufficiently deep pool. The ambient temperature was carefully controlled to 20 °C in every experiment.

Results

X-ray imaging experiments. We used X-ray imaging experiments equipped with a drop-impact setup, as illustrated in Fig. 1(a). The experiments were conducted at XSD 32-ID undulator beamline of the Advanced Photon Source in the Argonne National Laboratory. The air entrainment during drop impact on a liquid pool, as depicted in Fig. 1(b), was successfully taken as shown in Fig. 1(c). The imaging speed of the camera was synchronized to the X-ray beam using delay generators, enabling us to capture images with period of 3.68 μs and exposure time of 472 ns.

To investigate the effects of liquid viscosity (\(\mu\)) and surface tension (\(\gamma\)), we tested two model systems: i) alkane oils (heptane (C7H16), dodecane (C12H26), and pentadecane (C15H32)) and ii) mixtures of water and glycerol (W\(_x\)-G\(_y\)) where \(x\) and \(y\) denote mass fractions; 0, 0.2, 0.4, 0.6, 0.8, and 1.0. The liquid properties at temperature \(T = 293\) K are summarized in Table 1, where all data were retrieved from the literature.

Table 1. Properties of liquids at \(T = 293\) K. Here \(x_G\) refers water-glycerol mixtures with a fraction of glycerol. All data were retrieved from the literature.

| Liquids     | \(\rho\) (kg m\(^{-3}\)) | \(\mu\) (mPa s) | \(\gamma\) (mN m\(^{-1}\)) |
|-------------|-----------------------------|------------------|-----------------------------|
| Heptane     | 683.59 (ref. 39)            | 0.415 (ref. 30)  | 21.5 (ref. 31)              |
| Dodecane    | 746.4 (ref. 35)             | 1.500 (ref. 36)  | 25.6 (ref. 37)              |
| Pentadecane | 768.3 (ref. 39)             | 2.841 (ref. 38)  | 27.12 (ref. 33)             |
| x\(_G\) = 0 | 998.0 (ref. 27)             | 1.005 (ref. 25)  | 72.0 (ref. 26)              |
| x\(_G\) = 0.2 | 1051.2 (ref. 27)           | 1.742 (ref. 25)  | 69.5 (ref. 26)              |
| x\(_G\) = 0.4 | 1100.4 (ref. 27)           | 3.685 (ref. 25)  | 67.9 (ref. 26)              |
| x\(_G\) = 0.6 | 1157.6 (ref. 27)           | 10.911 (ref. 25) | 66.9 (ref. 26)              |
| x\(_G\) = 0.8 | 1210.7 (ref. 27)           | 60.856 (ref. 25) | 65.7 (ref. 26)              |
| x\(_G\) = 1.0 | 1263.9 (ref. 27)           | 1413.8 (ref. 25) | 62.5 (ref. 26)              |
| x\(_G\) = 0.6 | 1215.5 (ref. 27)           | 60.856 (ref. 25) | 65.7 (ref. 26)              |
| x\(_G\) = 1.0 | 1263.9 (ref. 27)           | 1413.8 (ref. 25) | 62.5 (ref. 26)              |

Figure 2 representatively shows sequential X-ray images that demonstrate the evolution of the entrained air in drop impact on a liquid pool for alkane liquids with small \(\gamma\) (20 ~ 27 mN/m). The external shape as well as the exact internal morphology of the entrained air are clearly visualized in high spatial resolution (~2 μm). The evolution from initial air films to final bubbles seems very elegant but quite complicated to understand. For the pentadecane in Fig. 2(a) and Movie S1, the air film initially evolves into a pancake-shape, is stretched into a vertical column at 118 μs, finally becomes one bubble at 177 μs. For the dodecane in Fig. 2(b) and Movie S2 with a lower viscosity, a longer vertical air column at 96 ~ 110 μs is formed and split into a double bubble (two bubbles) at 133 μs. For the heptane in Fig. 2(c) and Movie S3 with a much lower viscosity, the evolution becomes more complicated as follow. The central region of a pancake-like air film is punctured during retraction, forming a toroidal shape at 59 ~ 66 μs. The vertical stretching of the toroidal bubble at 66 ~ 81 μs induces the pinch-off of a daughter droplet at 81 ~ 88 μs.
finally encapsulated by an air shell, forming an antibubble. The antibubble seems to have a similar configuration with a typical antibubble, but the generation principle is quite different. Whereas the surfactant-stabilized antibubble is formed by oriented surfactant molecules that provide some elasticity to the air/liquid interfaces and ensure air-shell stability, the origin of the surfactant-free antibubble is the entrapped air by drop impact. These results clearly demonstrate that generation of single or double bubbles and antibubbles can be controlled during drop impact on a liquid pool by simply manipulating liquid viscosity.

For a water-glycerol mixture with large $\gamma$ (62 – 72 mN/m), we observed a similar viscosity-dependent air evolution, as demonstrated in Fig. 3. During retraction, the air evolved into one bubble for the high-viscosity $W_{0.8}G_{0.6}$ as shown in Fig. 3(a) and Movie S4 but split into a double bubble (two bubbles) for the intermediate-viscosity $W_{0.6}G_{0.4}$ in Fig. 3(b) and Movie S5 as well as $W_{0.8}G_{0.2}$ in Fig. 3(c) and Movie S6. For the low-viscosity pure water, a daughter droplet was formed in Fig. 3(d) and Movie S7 but the toroidal bubble was eventually split at 96 – 111 $\mu$s into a double bubble. Here we note that the initial retracting discs show undulation (red arrows in Fig. 3) and tiny bubbles were formed in the later stages (yellow circles in Fig. 3).
The retraction of the entrained air film in drop impact on a liquid pool has been studied mostly in inviscid liquids. Here, we systematically investigated retraction dynamics in a wide range of liquids. The retraction dynamics of a thin fluid sheet is governed by the competition among inertia, capillarity, and viscosity. As plotted in Fig. 5, we obtain a phase diagram for the final fate of the air as functions of the inverse Ohnesorge number (Oh) and viscosity. As defined as \( \frac{\tau}{\tau_i} \approx 0.5 \) for Oh \( > 10 \), the retraction rate significantly deviates from the exponential decay. The best-fit values of the proportional coefficients \( C_i \) and \( C_v \) are almost invariant (1.3 ~ 1.8) when Oh \( < 0.1 \) and aqueous solutions (\( C_i \approx 1.8 \)) were found at the inertial regime (Oh \( < 0.1 \)). This can be explained by the hydrodynamic instability by large surface tension as in aqueous liquids. Necklace rims are formed in aqueous liquids in Fig. 3, while smooth rims are formed in alkane liquids in Fig. 2. The necklace rim is responsible for the formation of tiny bubbles (dashed circles in Fig. 3) attributed to the capillary instability along the rim. Interestingly, a slight difference in \( C_i \) for oils (\( C_i \approx 1.8 \)) and aqueous solutions (\( C_i \approx 1.3 \)) was found at the inertial regime (Oh \( < 0.1 \)). This can be explained by the hydrodynamic instability by large surface tension as in aqueous liquids.
found at low Oh$^{-1}$ and low We$_{\text{film}}$ (blue region) and a split of one into two bubbles is found at low Oh$^{-1}$ and high We$_{\text{film}}$ (red region). At high Oh$^{-1}$ (green region), a daughter droplet is robustly formed but all three cases of one and two bubbles, or a shell bubble are possible.

The bubble breakup can be explained with respect to the competition among inertia, capillarity, and viscosity on the bubble columns vertically elongated caused by the inertia of retraction, as seen in Figs. 2 and 3. This inertia of retraction, or retraction speed, is proportional to the impact speed of liquid drop because the initial thickness of air film as well as the size of trapped air bubble become smaller with the impact speed. Thus, as seen in the inset of Fig. 5, the aspect ratio $L_o$ of the bubbles, measured at their maximum elongation (for example, at $t = 96 \mu s$ in Fig. 2(c)), increases with the kinetic energy of impacting liquid drop as $L_o \sim \text{We}_{\text{film}}^{\beta}$, where the best-fit exponent $\beta$ is measured as $\sim 0.486 \pm 0.037$. We find that the breakup usually occurs at high aspect ratios larger than a critical value, $L_o \sim 3.1$ in our data. The critical aspect ratio for typical breakup of fluid columns increases with fluid viscosity. From the relations of $L_o \sim \text{We}_{\text{film}}^{\beta}$ and $L_o \sim \text{Oh}^{1/2}$, the dependence of the critical We$_{\text{film}}^*$ on Oh can be rationalized as $\text{We}_{\text{film}}^* \sim \text{Oh}^{1/2 \beta}$. Therefore, the daughter droplet can be formed for low-viscosity liquids with Oh$^{-1} > 40$ (Oh < 0.025) in Fig. 5. Additionally,
decreasing Oh at Oh < 0.03 would increase the daughter droplet size, as seen in the ratio of the daughter droplet volume ($V_D$) to the bubble volume ($V_B$) measured as a function of Oh in Fig. 6(b). The pinch-off driven formation of the daughter droplet at Oh $-1 > 40$ or Oh $< 0.025$ is consistent between the theory and the experiment in Figs. 5 and 6(b).

The interaction of a daughter droplet with the surrounding liquid plays a crucial role in determining the final fate of the bubble. For the heptane, the daughter droplet coalesces axisymmetrically with the surrounding liquid and splits the bubble into a double bubble (Fig. 7(a)) or does not coalesce and instead remains inside the shell bubble, forming an antibubble (Fig. 7(b)). In rare cases, the daughter droplet forms one-sided coalescence with the surrounding liquid, resulting in one bubble (Fig. 7(c)). For water, the daughter droplet forms total coalescence (Fig. 8(a)) or one-sided coalescence at the equator of the vertically elongated bubble (Fig. 8(b)). In some events, the daughter droplet can coalesce with the surrounding liquid at the bottom of the bubble, resulting in one bubble as well (Fig. 8(c)). The coalescence is a random process and may not always happen in the center. For water, there was no event of shell bubble formation in our experiments.

The frequency of all the cases of single, double, or anti-bubbles at various experimental conditions are plotted in Fig. 9(a). The radius of the final spherical bubble $R_f$ is the average radius of the final single, double, or anti-bubbles, taken from the final bubble images. In a view of typical hydrodynamics, the air layer between surrounding...
liquid and the daughter droplet becomes very thin (< 1 μm) after the pinch-off. Thus, the layer is easily broken in water because of its high disjoining pressure, resulting in the split of the bubble into two, as illustrated in Fig. 9(b). Conversely, the thin air layer is not easily broken in oil because of its small surface tension, thus playing as a lubricating layer between the daughter droplet and surrounding oil, as illustrated in Fig. 9(c). Additionally, hydrodynamic instability may induce one-sided coalescence of the daughter droplet, eventually forming one bubble, as illustrated in Fig. 9(d). One interesting point in Fig. 9(a) is that for water, the events of no-split for one bubble are more frequently observed at high impact heights. This can be explained by the decrease in the bubble size with impact height, as shown in the top panel of Fig. 9(a). The increase in Oh by the decreased bubble size results in the decrease of the daughter droplet size, reducing the coalescence probability.

Discussion

The speed of impacting drop can significantly affect the initial dynamics of air. When the speed is low as $U < 0.5$ m/s, rupture occurs at numerous locations simultaneously, entrapping a multitude of tiny bubbles, known as the Mesler entrainment\(^{18,50}\). For $U > 0.5$ m/s, the air layer under the drop ruptures along an azimuthal ring and a disc-shaped air film is stably entrapped\(^{18,20}\). Especially, for $U = 1.0 \sim 2.5$ m/s, as applied here, the evolution dynamics of the air films is hardly affected by the impacting speed, except for their size dependency on the speed\(^{19}\), as fitted by $R_B \sim U^{-0.436}$ in Fig. 10. This relation estimates $R_B \sim 70 \mu m$ at $U \sim 0.25$ m/s, as consistent with the previous measurement of $R_B \sim 60 \mu m$\(^{51}\) for an ethanol droplet of 0.9 mm radius at $U \sim 0.25$ m/s.

Here, the Weber number of the impacting drop was markedly similar between this study for drop impact on liquid pools ($We_{drop} \sim 50 \sim 210$) and previous studies for drop impact on solid substrates ($55 \sim 70$ or $70 \sim 900$), but the below boundary condition was markedly different. This study clearly reveals the variety of air evolution dynamics. At very high impact speeds, collapsing of the impact crater would be a main mechanism for the bubble entrainment\(^{16}\), generally known as regular bubble entrainment. Also, unusual phenomena such as the cascade entrapment of bubble rings\(^{21}\), and the formation of vortex streets also can occur\(^{52}\). By expanding the experimental conditions, one may be able to explore such singular hydrodynamic behavior in drop impact on liquid using novel techniques such as ultrafast X-ray phase-contrast imaging.

Finally, we did not consider the effect of variation of air properties such as air temperature and viscosity. The change in air temperature can modify the properties of liquid, in particular liquid viscosity (generally, the viscosity becomes smaller at a higher temperature), and thus can shift the boundaries between regions in the phase diagram. The change in air viscosity significantly affects the whole dynamics of drop impact and thus it would be crucial to investigate the effect of air viscosity on air entrainment during drop impact on a liquid pool. Further studies should be expanded to understand the oblique impact of droplets on deep liquid pools\(^{24-30}\), the impact of viscoelastic or viscous droplets\(^{27-39}\), and the impact-induced fabrication\(^{40-44}\), with more facilitating of numerical\(^{35}\), theoretical\(^{36}\), and experimental approaches\(^{45,46}\).
Conclusion

In summary, the novel ultrafast X-ray phase-contrast imaging is useful for us to explore the dynamics of the air entrainment during drop impact on a liquid pool. We elaborated two retraction mechanisms of the entrapped air film, inertial and viscous retractions, which strongly depend on the liquid viscosity. We found two crucial dynamic singularities after retraction: (i) the breakup of the bubble, mostly characterized with the bubble geometry and the liquid property, and (ii) the pinch-off of the daughter droplet (at Oh < 0.025) and its effect on the final fate of the bubble, which have been unachievable using conventional imaging techniques. Finally, predicting the morphological evolution of the entrained air would be assessable via the phase diagram with respect to hydrodynamic conditions.

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Author contributions
J.S.L., B.M.W. and J.H.J. designed the study. J.S.L. and S.J.P. conducted the main experiments. J.S.L. and B.M.W. collected the data and analyzed the results. J.T.K. and J.P. helped in the experiments. K.F. managed the X-ray imaging facility. J.H.J. supervised the study. J.S.L., B.M.W. and J.H.J. wrote the initial manuscript. B.M.W. and J.H.J. revised the final manuscript. All authors discussed the results.

Competing interests
The authors declare no competing interests.

Additional information
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