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Are Drop-Impact Phenomena Described by Rayleigh-Taylor or Kelvin-Helmholtz Theory?

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Drop impact, spreading, fingering, and snap-off are important in many engineering applications such as spray drying, industrial painting, environmentally friendly combustion, inkjet printing, materials processing, fire suppression, and pharmaceutical coating. Controlling drop-impact instability is crucial to designing optimized systems for the aforementioned applications. Classical Rayleigh-Taylor (RT) theory has been widely used to analyze fingering where instabilities at the leading edge of the toroidal ring form fingers that may ultimately snap off to form small droplets. In this study, we demonstrate the inapplicability of RT theory, in particular because it fails to explain the stable regimes observed under conditions of low air density and the instabilities observed when a drop impacts a pool of equal-density fluid. Specifically, finger instability decreases with decreasing air density, whereas the RT theory suggests that instability should remain unchanged. Moreover, experiments show that fingers form upon impact of a dyed water drop with a water pool, whereas the RT theory predicts no instability when the densities of the two interacting fluids are equal. Experimental evidence is instead consistent with instability predictions made using the shear-driven Kelvin-Helmholtz theory.

Keywords Drop impact; Finger instability; Kelvin-Helmholtz; Rayleigh-Taylor; Splash

INTRODUCTION

Engineers, scientists, and the general public have been fascinated by the nebulous beauty of drop spreading and snap-off for more than a century. One of the first relevant pioneering scientific works was carried out by Worthington^[1] nearly a century ago and since then, interest in drop-impact phenomena has continued because of its practicality in numerous engineering applications such as spray drying,^[2–6] industrial painting, environmentally friendly

combustion,^[7–10] inkjet printing, materials processing, and pharmaceutical material coating.^[11–13] It is also relevant to spray-drying applications where powders are produced from suspensions and solutions using various atomizers. In these applications, understanding drop-impact phenomena is important if the goal is to minimize deposits on spray-dryer walls or to produce powders by spraying monodisperse drops onto various substrates. A consistent predictor of fundamental instabilities (that may ultimately lead to snap-off) is needed to support technological advancement in the aforementioned applications.

Previously, the properties of the falling fluid and impacting substrate were thought to be the primary parameters controlling the instabilities that lead to snap-off. The drop and substrate can both be either solid or liquid; cold or hot; miscible or immiscible; rough or smooth; chemically reacting or nonreacting; and Newtonian or non-Newtonian.^[14] Fingers at the spreading liquid edge/interface in the azimuthal direction at the cusp of the toroidal sheet of rising liquid (the “corona”) and subsequent splashing (“ejection” or “snap-off”) are often formed when the Weber number, defined as $We = \rho DU^2 / \sigma$, is sufficiently high. We is the ratio of inertial to surface-tension forces of a drop where ρ , D , U , and σ are the drop density, diameter, impact speed, and surface tension, respectively. Experimental data demonstrate, however, that a sufficiently high We to form fingers under high ambient pressure may be insufficient in systems with low ambient pressures. Fingering is a necessary but not sufficient condition for splashing (or snap-off). That is, there are certainly many instances of fingering without snap-off, but the reverse is never observed.

Fingering must be a response to some disturbance during droplet impact.^[15] This immediately leads to the question: What is the source of the disturbance? Previously, acceleration-driven Rayleigh-Taylor (RT) instability was applied to predict the dominant wavelength (number of

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fingers) forming along the corona of an impacting drop.^[16–18] During the collapse of a liquid drop, the spreading rate increases with time (accelerates) until the top of the collapsing drop contacts the substrate.^[19] Afterward, the spreading decelerates due to viscous dissipation and ambient air resistance. The commonly held theory has been that this acceleration/deceleration cycle is the source of RT instability.^[16]

In this study, we present overwhelming experimental evidence that demonstrates how RT theory fails to describe the complete spectrum of fingering observations, specifically those occurring in systems with minimal atmosphere or when drop and liquid substrate densities approach equality. On the other hand, Kelvin-Helmholtz (KH) instability theory^[20] explains not only the general trends but the limiting cases observed in previously published and these new experimental data. From a more practical standpoint, we also establish the critical Weber number and ambient pressure (We_{crit} and P_{crit}) where instabilities are first observed.

EXPERIMENTAL METHODS

The liquids used in these experiments are C_6F_{14} (FC72) Fluorinert (a refrigerant fluid surrogate for hydrocarbon fuels and cryogenic liquids), water, and ethanol. A precision microliter valve (Model 740V-SS, EFD Inc., East Providence, RI) with stainless steel tips was used to generate 1.5 to 4 mm diameter drops released from heights of up to 1.32 m to obtain impact speeds of up to 5 m/s. A variable-pressure, aluminum chamber equipped with two polycarbonate windows for illumination and photography housed experiments carried out at pressures between 0.2 and 6 atm. For most tests, the impact surface is smooth Plexiglas ($<8\mu\text{m}$ roughness). Drop temperature was 298 K to maintain constant fluid properties and eliminate variability due to evaporative cooling.^[21,22] For impact tests on a water pool, dyed water drops were used. Impact and spreading sequences were captured using a Vision Research Phantom V9 high-speed video camera (4,800 to 17,021 fps).

Evaluation of Instability Theories

The question is naturally asked: What is the source of the instability that leads to fingering and snap-off? In addition to RT instability theory, others have proposed Plateau-Rayleigh (PR or surface tension-driven) capillary instability^[23] as the source of hydrodynamic instability leading to fingering. Here, RT, PR, and KH^[24] instability theories are all assessed with regard to their applicability across a range of system parameters and limiting conditions. We demonstrate that only KH theory successfully describes the complete spectrum of experimental observations.

Background

Before embarking on our mission to identify sources of instability, first we investigated the effect of ambient air pressure. Pressure is directly correlated to density and Table 1 lists the critical air pressures where instabilities or fingers are first observed during drop spreading ($P_{crit,1}$) of FC72, water, and ethanol on Plexiglas. These critical pressures imply that air pressure plays a pivotal role in the formation of instabilities; as air pressure decreases, instabilities disappear. For general interest, the second critical pressure, $P_{crit,2}$, is where fingers are first observed to snap off into droplets.

The experimental image in Fig. 1 (left column) demonstrates the appearance of fingers at $We = 695$. While reducing air pressure suppresses instability, low impact speeds (or low We) have the same effect because of the stabilizing force of surface tension, as evident in the experiments at $We = 50$ also shown in Fig. 1 (right column).

An alternate hypothesis is presented: As an impacting drop spreads, the no-slip boundary condition and viscous forces build a boundary layer with an adverse pressure gradient (a fundamental source of instability that may lead to turbulence) that would tend to lift the fluid from the substrate. An unstable mode might be triggered in such a system and could be the source of instability leading to fingering. However, this hypothesis is not born out by the experiments^[15] because at a constant We , fingers do not appear under low air pressure; see Fig. 2. It seems that the shear effects of air are more important in splashing phenomena that is an adverse pressure gradient.

Evaluation of Rayleigh-Taylor Instability Theory

Traditionally, the fluid dynamics community has accepted the notion that finger formation is caused by acceleration-driven RT instability.^[16]

$$\omega = \frac{\rho - \rho_s}{\rho + \rho_s} a_c k - \frac{\sigma k^3}{\rho + \rho_s}, \quad (1)$$

where a_c is the acceleration of the interface from gas to liquid, σ is the liquid surface tension, ρ_s is the surrounding

TABLE 1
The critical air pressures for FC72, water, and ethanol drops

	We	$P_{crit,1}^a$ (atm)	$P_{crit,2}^b$ (atm)	ρ (kg/m^3)	σ (kg/s^2)
FC72	955	0.50	0.90	1680	0.012
FC72	1970	0.30	0.70	1680	0.012
Water	1000	0.20	0.80	1000	0.073
Ethanol	1675	0.30	0.40	790	0.022

^aFinger formation.

^bFinger snap-off.

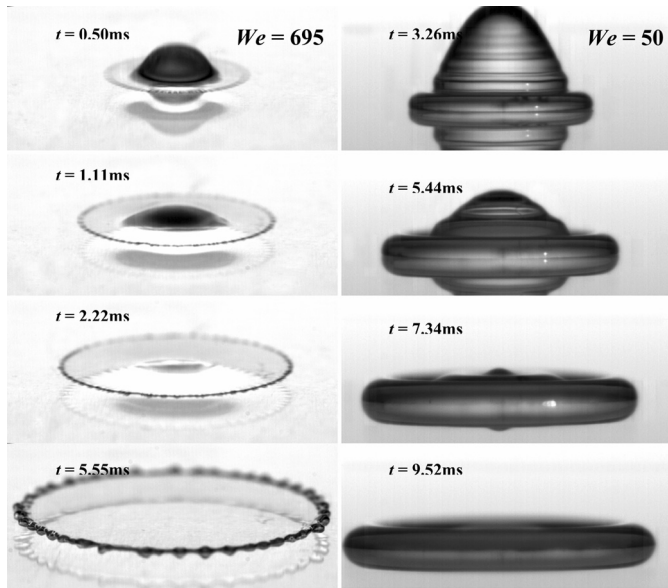


FIG. 1. Photographic series (from top to bottom) for a water drop impact onto Plexiglas at 1 atm. The left column ($We = 695$) was captured from a higher camera angle to illustrate finger formation and the right column ($We = 50$) was captured with the camera perpendicular to the impact surface.

gas density, and ω is the complex growth rate comprising both the real finger growth rate and the imaginary frequency of oscillation (number of fingers). Upon applying $d\omega/dk = 0$, (1) yields the dominant wave number (k_{\max}):

$$k_{\max} = \sqrt{\frac{a_c(\rho - \rho_s)}{3\sigma}}. \quad (2)$$

The preceding equation indicates a strong unstable mode, particularly when the surrounding gas density tends to

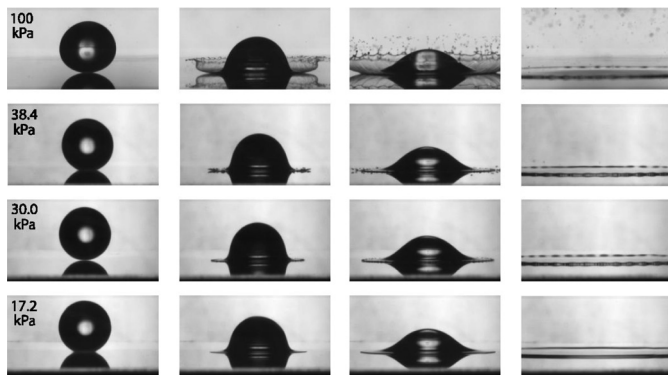


FIG. 2. Recent experiments by Xu et al.^[3] of the University of Chicago. Fingers were not observed when the ambient air pressure decreased. Also note that the drop is most deformed (oblate spheroid) under highest ambient air pressure due to compressed, escaping air. Reprinted with the permission of Prof. Sidney R. Nagel of University of Chicago.

zero. This is clearly contrary to our experiments and those of Xu et al.,^[15] which always show a stable mode ($k_{\max} \rightarrow 0$ or no finger formation) as the air density is reduced ($\rho_s \rightarrow 0$); see Fig. 2.

To further investigate the RT-based instability hypothesis, additional tests with dyed water drops impacting a liquid (water) pool were conducted at $We = 50$ as shown in Fig. 3. Here, given the relatively low We , no rising sheet or snap-off is induced above the pool's surface; only radial wave propagation is observed in Fig. 3(a). However, the bottom view in Fig. 3(b) shows obvious finger formation in the pool. Clearly, coronal fingering of a liquid drop on Plexiglas resembles that in the liquid pool after drop impact. Because there is no capillary effect at the interface due to fluid cohesion, PR capillary instability cannot exist. Furthermore, RT instability cannot be responsible for this finger formation because there is no density gradient (the drop density is the same as the surrounding density and $\rho = \rho_s$). Because the second term on the right-hand side of (1) indicates that the growth rate is negative when $\rho = \rho_s$, RT theory suggests that the system is unconditionally stable. Because fingers indeed appear at the interface, RT instability theory cannot be used to explain this scenario. The only remaining destabilizing mechanism in the system is the shear gradient.

At sufficiently high We , coronal azimuthal waves form upon impact (see the snapshot at $t = 0.2$ ms in Fig. 4). At this early stage, the length scales of the waves (which eventually become fingers) are relatively small, a limiting case of $\lambda/\ell \rightarrow 0$ (i.e., where λ and ℓ are the most dominant unstable wavelength and the characteristic length scale, respectively). While the impact process progresses, the number of fingers around the corona decreases as they merge and small-scale disturbances are damped out. As seen in Fig. 4, there are numerous fingers when $t < 6.0$ ms, but later, fingers merge as capillary and cohesive forces begin to manifest themselves. Could this be PR instability?

Evaluation of Plateau-Rayleigh Instability Theory

Taylor^[25] and Rieber and Frohn^[26] proposed that finger formation is due to PR capillary instability,^[23] but

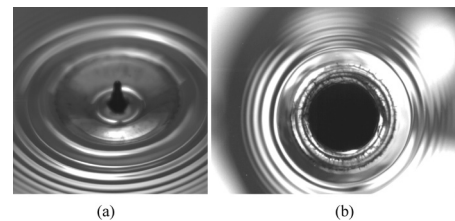


FIG. 3. The 2-mm dyed water drop was released from a height of 0.092 m with the impact speed of $U = 1.34$ m/s; $We = 50$, $p = 1$ atm. These photos were taken 43 ms after initial impact. (a) Angled view; (b) bottom view.

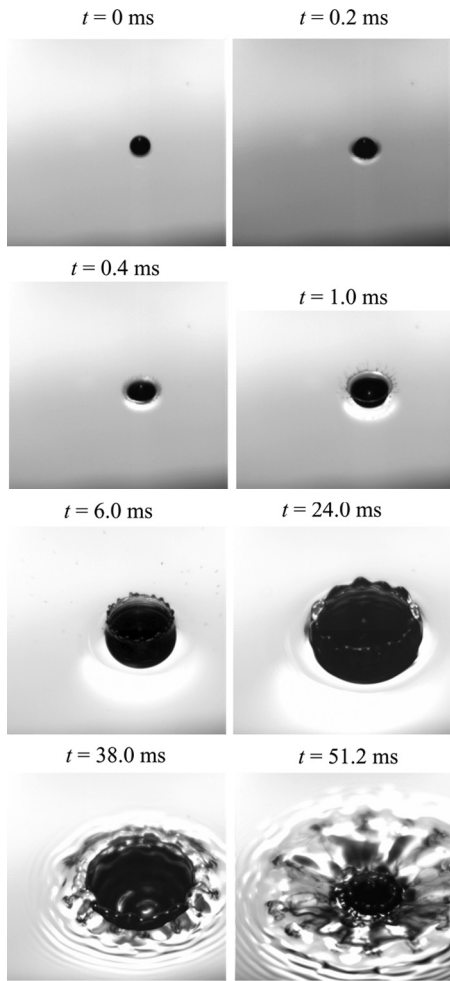


FIG. 4. Time series snapshots of a drop impact onto a water pool. The 2-mm dyed water drop was released from a height of 0.3 m yielding $We = 163$, $p = 1$ atm.

this was refuted by Yarin,^[14] who stated that PR mechanisms significantly underpredicted the observed number of fingers. Yarin also noted that finger formation in Rieber and Frohn's numerical simulation was only possible because of their artificially specified disturbance; the fingers would not form without this. Fullana and Zaleski^[27] also studied coronal instability processes. By growing a span-wise sinusoidal perturbation, they numerically demonstrated that only oversized waves (fingers) appeared at the rim, contrary to experimental observations.^[1,28] Even with sufficiently long simulation time, their oversized fingers grew much too slowly and never resulted in breakup, unlike observations of a cylindrical liquid jet subject to PR instability. Finally, they ruled out the possibility of PR instability causing finger formation and suggested that some other destabilizing force must exist but speculated no further. Though the notion of PR capillary instability causing finger formation does not seem to be entirely

wrong, it appears that an additional destabilizing force is needed to generate the small azimuthal wavelengths initially observed.

As Fig. 4 illustrates, the unstable wavelength early in the impact sequence is much smaller than observed at later stages. Small- and large-wavelength fingers are likely governed by distinct mechanisms, each showing dominance at various stages in the impact sequence. There must be some initial and more fundamental instability (beyond PR instability) that promulgates the many fingers (e.g., 21 fingers at 6 ms), which are later impacted by capillary and cohesive forces that merge fingers and increase observed wavelengths (e.g., 13 fingers at 24 ms). It seems unfathomable that the numerous small fingers observed for the drop-pool impact is due to PR instability because the fluids are identical and surface tension-driven instability should be essentially absent. Could KH instability arise from the significant shear stresses expected for near-equal-density fluid impacts?

Evaluation of Kelvin-Helmholtz Instability Theory

Hydrodynamic stability analysis dates back to the work of Lord Rayleigh,^[23] who developed PR instability theory; since then, many have succeeded him.^[29–34] Yoon and Heister^[24] showed that all equations of the aforementioned authors are equivalent for inviscid flows with relatively small length scales (i.e., the limit of $\lambda/1 \rightarrow 0$ or $k1 \rightarrow \infty$, with $k = 2\pi/\lambda$). For the inviscid case, all equations reduce to the KH dispersion relation, which, when neglecting gravity, reduce to the liquid sheet instability of Taylor.^[25] These KH (or Taylor) equations assume that surface tension and inertial forces are the major competing forces:

$$\omega^2 = \frac{\rho_s}{\rho} U_{diff}^2 k^2 - \frac{\sigma}{\rho} k^3. \quad (3)$$

where U_{diff} is the velocity difference between the incoming drop and the escaping air. Equation (3) is derived under the assumption that ρ_s/ρ terms are negligible, except for the term containing U_{diff}^2 where the shear effect is most pronounced (Levich^[32] provides the most general form of the dispersion equation that includes all terms). The dominant wave number obtained when solving $d\omega/dk = 0$ is

$$k_{max} = \frac{2\rho_s U_{diff}^2}{3\sigma}, \quad (4)$$

The characteristic length scale, ℓ , does not appear in (3) or (4) because the wavelength of interest, λ , is extremely small, compared to 1; all terms involving $\lambda/1$ were taken in the limit of $\lambda/1 \rightarrow 0$. We postulate that the initial disturbance leading to finger formation can be fully explained in terms of shear-driven KH instability theory.

Support for this hypothesis is gained through analysis of (4), which indicates that in the absence of air (i.e., $\rho_s = 0$), $k_{\max} \rightarrow 0$ instabilities are suppressed, consistent with the experimental observations of Xu et al.^[15] Note that for such a system ($\rho_s = 0$), RT theory in (2) predicts a finite number of fingers, $k_{\max} \rightarrow \sqrt{a_c \rho / 3\sigma}$, which is clearly counter to experimental evidence. At the other end of the spectrum, when the drop and surrounding fluid density are the same (i.e., $\rho_s \rightarrow \rho$), KH theory predicts a dramatic increase in the wave number consistent with the pictures in Figs. 3 and 4, while RT theory predicts no fingers ($k_{\max} \rightarrow 0$).

Figure 5 is the snapshot taken from below the drop-pool impact at 42 ms for the same event shown in Fig. 4. The 13 fingers below the pool surface equate to the 13 anti-waves observed along the coronal rim at 24 ms in Fig. 4. This demonstrates that fingering occurs even for equal-density fluids and that they retain the negative imprint of the initial shear perturbation from the escaping air. Recall from Fig. 4 that it is the escaping air that provides the initial instability governing finger formation when We is sufficiently high (here $We = 163$). Recall also that at relatively low We ($We = 50$ as in Fig. 1), shear disturbance from the air is insufficient to initiate finger formation in the coronal ring. Nevertheless, in Fig. 5, the shear disturbances at the drop-pool interface apparently provide the instability mechanism that forms fingers as the drop spreads into the pool (Fig. 3(b)).

Based on the experimental evidence presented herein, we conclude that fingers are formed from shear instabilities between the liquid and the air escaping from between the falling drop and a solid impact substrate. For the drop-into-pool experiments, it is also the gradient of liquid-liquid shear that elicits finger formation. The source of instability is much better explained by shear-driven KH

theory, which suggests that it is the perturbation of the surface layer of the spreading drop that induces finger formation at the periphery of the spreading liquid. Moreover, acceleration-driven RT theory cannot be used to describe the source of instabilities that lead to fingering or we would have observed many fingers in low ambient pressure systems and none for an impacting drop spreading within an equal-density liquid pool. In fact, the opposite was observed in both limiting cases.

CONCLUSIONS

This work provides a new physical framework describing the source of instabilities that leads to finger formation upon drop impacts on various substrates. Finger formation is clearly a function of both We and ambient air pressure for impacting drops. Two different types of experiments clearly demonstrate that instability is not described by acceleration-driven RT theory or surface tension-driven PR theory, but instead by shear-driven KH theory, which is well suited for describing drop impact onto both a smooth substrate and a liquid pool. Experiments for drops on Plexiglas demonstrate that finger formation can be predicted based on the combination of We and ambient air pressure. Drop-pool impacts also manifest the effects of a shear disturbance, which has imprinted its instability on the drop's subsequent spread into the pool. Clearly, it is important not to neglect shear effects from the surrounding air or liquid pool on drop impact characteristics. The implications of these results provide guidance to engineering processes such as spray drying, fuel dispersion and ignition, ink jet printing, painting, materials processing, and pharmaceutical coating applications where KH theory can be used to predict both initiation of fingers and the number of fingers expected upon drop impacts.

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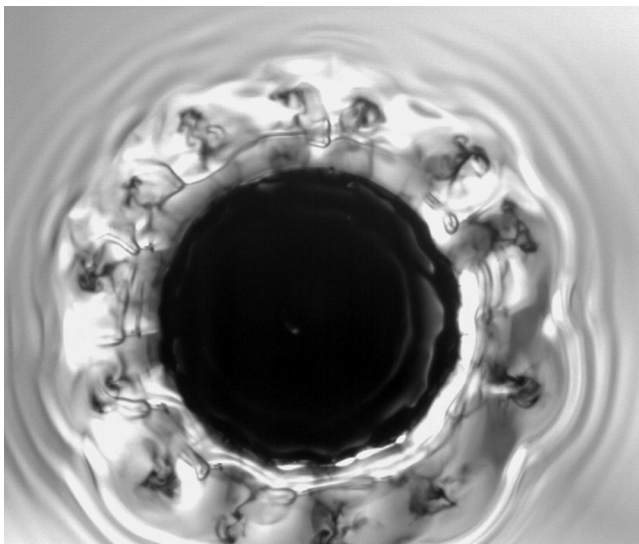


FIG. 5. A snapshot of the bottom view of Fig. 4 at 42 ms.

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