Instability development of a viscous liquid drop impacting a smooth substrate

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Keywords: impact, instability, air pressure, viscosity

We study the instability development during a viscous liquid drop impacting a smooth substrate, using high speed photography. The onset time of the instability highly depends on the surrounding air pressure and the liquid viscosity: it decreases with air pressure with the power of minus two, and increases linearly with the liquid viscosity. From the real-time dynamics measurements, we construct a model which compares the destabilizing stress from air with the stabilizing stress from liquid viscosity. Under this model, our experimental results indicate that at the instability onset time, the two stresses balance each other. This model also illustrates the different mechanisms for the inviscid and viscous regimes previously observed: the inviscid regime is stabilized by the surface tension and the viscous regime is stabilized by the liquid viscosity.

PACS numbers: 47.20.-k, 47.20.Ma, 47.55.Ca, 47.55.dr

The phenomenon of a liquid drop hitting a solid surface is ubiquitous: it occurs whenever the very first rain drop reaches the ground or when we spill coffee onto the floor. Liquid-solid impact has been extensively studied due to its broad applications in many industrial processes, such as ink-jet printing, surface coating, combustion of liquid fuel, plasma spraying, and pesticide application[1]. It may seem obvious that the impact outcomes should be determined by either the liquid or the solid properties[2-7], however, recent studies surprisingly revealed the crucial role of the surrounding atmosphere: reducing air pressure can completely suppress the liquid drop splashing on a smooth substrate[8,9], and the compressibility of the surrounding air is demonstrated to be important[10,11]. This unexpected discovery brings a completely new effect, the air effect, into the impact problem. To fully understand this new effect, therefore, it is essential to clarify the interactions between air and the fundamental liquid properties, such as surface tension and viscosity. Previous study has shown that the competition between the air effect and the liquid surface tension determines the impact outcomes of inviscid liquid drops[8]. However, there has been very limited study on the interaction between air and the liquid viscosity, although the liquid viscosity itself has been broadly tested[2,3,12] and the entrainment of air bubbles in viscous drops was illustrated recently[13,14]. As a result, the relationship between surrounding air and the liquid viscosity is still missing, and the understanding on the liquid-solid impacts, especially the newly discovered air effect, remains incomplete.

In this paper, we systematically study the interaction between air and the liquid viscosity by varying both the surrounding air pressure and the liquid viscosity, for the impacts of viscous liquid drops on a smooth substrate. With high speed photography, we find that the instability produced by an impact highly depends on the air pressure and the liquid viscosity: the onset time of the instability decreases with air pressure with the power law of minus two, while it increases linearly with the liquid viscosity. From the real-time liquid motion measurements, we construct a simple model that compares the destabilizing stress from air with the stabilizing stress from the liquid viscous stress. The experimental results support the picture that the two stresses balance each other at the instability onset time. This model also predicts the existence of a threshold viscosity, above which the system is stabilized by the liquid viscosity, and below which it is stabilized by the surface tension. This prediction quantitatively agrees with the previous experiment[8].

We perform all the experiments inside a transparent vacuum chamber whose pressure can be continuously varied from 1kPa to 102kPa (atmospheric pressure). We also independently vary the liquid viscosity by using silicone oils of very close densities (0.92 ~ 0.94 g cm$^{-3}$) and surface tensions (19.7 ~ 20.5 mN m$^{-1}$) but different dynamic viscosities (4.65 ~ 13.2 mPa s). We note that all our liquids wet the substrate completely thus the wetting conditions are kept the same for all the impacts. To make sure that identical impact conditions are achieved each time, we release reproducible liquid drops of diameter $d = 3.1 \pm 0.1$ mm from a fixed height, and all the liquid drops impact a smooth and dry glass substrate at the velocity $V_0 = 4.03 \pm 0.05$ m s$^{-1}$. The impacts are subsequently recorded by a high speed camera at the frame rate of 47,000 frames per second.

We probe the air-liquid interaction by inspecting the instability development during the impact: under high-speed photography, the impact produces a thin liquid film expanding radially along the substrate. This liquid film is stable initially, however, a small rim shows up around the edge at a certain moment, and subsequently develops into larger and larger undulations (See Fig.1 left column). We believe the appearance of the rim indicates the transition from a stable system into an unstable one, and define the moment of the rim appearance as the instability onset time, $t_{on}$. For example, an instant very close to $t_{on}$ is shown in the third image of Fig.1 left column. This instability onset time, $t_{on}$, measures how fast the system goes unstable: the smaller it is, the faster the system becomes unstable. Interestingly, $t_{on}$ has a strong dependence on the surrounding air pressure, $P$. The two columns in Fig.1 show two almost identical impacts, with only different $P$: At $P = 40$ kPa (left column), instabilities show up in the third image; while at a higher pressure, $P = 63$ kPa (right column), they appear at a much earlier time in the second image.

By performing similar experiments under different air pressures, we systematically measure the instability onset time, $t_{on}$, with respect to the pressure, $P$. We find that $t_{on}$ decreases monotonically with $P$, as shown in Fig.2. Intuitively,
this implies that more air leads to earlier instability appearance, thus air acts to destabilize the system, consistent with previous findings\cite{8}. To test the interaction between air pressure and liquid viscosity, we perform the same measurements with silicone oils of very similar mass density and surface tension, but different dynamic viscosities, as plotted by the different symbols in Fig.2. From bottom to top, the four curves correspond to increasing dynamic viscosities: $\mu = 4.65(\bullet), 6.7(\circ), 9.3(\Delta)$, and $13.2(\times)$ mPa s. Intriguingly, all the data can be excellently fitted by a simple functional form: $t_{\text{on}} = A/P^2 + t_0$, with $A$ and $t_0$ the fitting parameters. $t_0$ has typical values between 0.03 to 0.09ms, much smaller than most $t_{\text{on}}$ values. However, it is still larger than our time resolution(0.02ms) and can not be explained as measurement errors. One possibility is that the system actually becomes unstable slightly earlier than the measured $t_{\text{on}}$, but the instability features at that moment are too tiny to visualize. The pre-factor, $A$, increases with the viscosity, $\mu$, as illustrated by the higher locations for larger $\mu$. Limited by the experimental condition, each data set has only about one decade in x and y directions. But it is nonetheless impressive that one simple functional form can fit all the data sets well.

Since $V_e$ and $d$ vary with time, so do $\Sigma_G$ and $\Sigma_\mu$. Therefore a careful examination on their time dependence could provide valuable insight for the instability development. We can directly measure $r(t)$ and $d(t)$ from high-speed photography, as illustrated in Fig.3 upper inset. $V_e$ can be obtained by taking the time derivative of $r(t)$. Our measurements show that $r(t) \propto \sqrt{t}$, consistent with previous studies, thus $V_e = dr/dt \propto 1/\sqrt{t}$. This time dependence keeps valid for most of the expanding period, within which all our measurements are performed. Moreover, we can directly measure the thickness of the liquid film, $d$, with respect to $t$, as plotted in the main panel of Fig.3. Because the small values of $d$ approach the single pixel level of our camera, the data are quite discrete; nonetheless they are consistent with the fit: $d \propto \sqrt{t}$, with $\nu = \mu/\rho_L$ being the liquid kinematic viscosity. This shows that $d$ is determined by the boundary layer thickness, $\sqrt{\nu t}$.

From the real-time dynamics, we derive the time dependence of the stresses: The destabilizing stress, $\Sigma_G \sim \rho_G \cdot C_G \cdot V_e \propto 1/\sqrt{t}$, decreases with $t$ with the power of $-\frac{1}{2}$;
while the stabilizing stress, $\Sigma_\mu \sim \mu V_e / d \propto 1/t$, depends on $t$ with the power of $-1$. Clearly, when $t$ is small, $\Sigma_\mu \gg \Sigma_G$, and the stabilizing stress dominates the destabilizing stress. This implies that the system should be stable initially, as we have observed. As $t$ increases, however, $\Sigma_\mu$ decreases much faster than $\Sigma_G$ and a crossover should occur at a certain time. After this crossover time, $\Sigma_G$ becomes dominant and the system will go unstable. The experiments are consistent with this picture: all the impacts are indeed stable initially and become unstable after the instability onset time, $t_{on}$. Therefore $t_{on}$ naturally corresponds to the crossover time at which the two stresses balance each other:

$$\rho G \cdot C_G \cdot V_e \sim \frac{\mu V_e}{d} \mid_{t=t_{on}}$$  \hspace{1cm} (1)

Plugging in the relations: $\rho G \propto P$ and $d \propto \sqrt{\mu} t$, with $V_e$ canceling each other on both sides and $C_G$ being a constant independent of $P$, we reach the expression:

$$t_{on} \propto \frac{\mu}{P^2}$$  \hspace{1cm} (2)

This expression successfully explains the two main features observed in Fig.2: (1) $t_{on} - t_0 \propto 1/P^2$ and (2) the pre-factor of this dependence, $A$, increases with $\mu$. Moreover, Eq. 2 further predicts that $A$ should increase linearly with $\mu$. To test this prediction, we find $A$ for each viscosity in Fig.2 from the best fit (the solid curves in Fig.2), and plot $A$ as the function of $\mu$ in Fig.4. Indeed, a very nice linear dependence is observed but the line does not go through the origin; instead, it intercepts the x-axis at the finite viscosity value, $\mu_0 = 3.4 \text{ mPa s}$.  

What is the physical meaning of $\mu_0$? To answer this question, we need to understand the impacts by the inviscid liquids with $\mu < \mu_0$. Previous study showed that for an inviscid liquid drop impacting on a smooth surface, the destabilizing stress is the same as the current viscid case, $\Sigma_G \sim \rho C_G V_e$. However, the stabilizing stress, $\Sigma_\mu$, is quite different. $\Sigma_\mu$ comes from the liquid surface tension, and is typically estimated as the surface tension coefficient, $\sigma$, divided by the liquid film thickness, $d$: $\Sigma_\mu \sim \sigma / d$. Therefore, we propose that the complete stabilizing effect for an impact should include both the surface tension component, $\Sigma_L$, and the viscosity component, $\Sigma_\mu$. When the viscosity is small, $\Sigma_L$ dominates $\Sigma_\mu$, and we get typical inviscid behavior. However, when $\mu$ exceeds a certain threshold value, the viscous stress $\Sigma_\mu$ will become the major stabilizing factor, and we get the currently observed viscous behavior. Therefore $\mu_0$ naturally corresponds to this threshold viscosity which determines whether the inviscid or the viscous model should be used. We note that $\mu_0$ should depend on detailed impact conditions such as the impact velocity, surface tension and wetting conditions. Previous experiments with similar impact conditions already confirmed that two impact regimes exist when $\mu$ is varied, and the transition from the inviscid regime to the viscous regime is close to $\mu_0$ (see ref. [9] Fig.5). This provides a strong experimental evidence for the physical meaning of $\mu_0$. Moreover, our picture not only explains the meaning of $\mu_0$, it also demonstrates the main difference between the two impact regimes: the inviscid regime is stabilized by the surface tension and the viscous regime is stabilized by the liquid viscosity.

We propose that in the viscous regime, the stabilizing stress is mainly from the viscous stress, $\Sigma_\mu \sim \mu V_e / d$, and construct a model which compares $\Sigma_\mu$ with the destabilizing stress, $\Sigma_G \sim \rho C_G V_e$. By assuming that $\Sigma_G$ and $\Sigma_\mu$ balance each other at the instability onset time, $t_{on}$ (Eq.1), we successfully explain the dependence of $t_{on}$ on $P$ and $\mu$: $t_{on} - t_0 = A/P^2$ and $A \propto \mu - \mu_0$, with $\mu_0$ the threshold viscosity separating the inviscid and the viscous regimes. However, the most critical criterion, whether $\Sigma_G$ and $\Sigma_\mu$ are indeed comparable at $t_{on}$, remains to be verified. To test it quantitatively, we measure the ratio between the two stresses, $\Sigma_G/\Sigma_\mu \sim \rho C_G d / \mu$, at

FIG. 3. Direct measurement of the thickness $d$ vs. time $t$. The impact is by a liquid drop of $\mu = 4.6 \text{ mPa s}$ and $V_e = 4.03 \pm 0.01 \text{ m/s}$. The inset shows a typical snapshot from which $d$ is measured: $d$ is the liquid film thickness measured at the edge. Main panel shows the measured $d(t)$. Because $d$ is quite small, the four discrete values correspond to one, two, three and four pixels of our camera. The fit is: $d = 1.9 \sqrt{\mu t}$, indicating that $d$ is determined by the boundary layer thickness: $\sqrt{\mu t}$.

FIG. 4. The pre-factor, $A$, vs. liquid viscosity, $\mu$, for the curves shown in Fig.2. The pre-factors are obtained from the best fits in Fig. 2. $A$ varies linearly with $\mu$ and intercepts the x-axis at $\mu_0 = 3.4 \text{ mPa s}$. $\mu_0$ agrees with the threshold viscosity separating the inviscid and viscous regimes observed in previous experiments.

The impact regimes: the inviscid regime is stabilized by the surface tension component, $\Sigma_L$, and the viscous regime is stabilized by the liquid viscosity component, $\Sigma_\mu$. When the viscosity is small, $\Sigma_L$ dominates $\Sigma_\mu$, and we get typical inviscid behavior. However, when $\mu$ exceeds a certain threshold value, the viscous stress $\Sigma_\mu$ will become the major stabilizing factor, and we get the currently observed viscous behavior. Therefore $\mu_0$ naturally corresponds to this threshold viscosity which determines whether the inviscid or the viscous model should be used. We note that $\mu_0$ should depend on detailed impact conditions such as the impact velocity, surface tension and wetting conditions. Previous experiments with similar impact conditions already confirmed that two impact regimes exist when $\mu$ is varied, and the transition from the inviscid regime to the viscous regime is close to $\mu_0$ (see ref. [9] Fig.5). This provides a strong experimental evidence for the physical meaning of $\mu_0$. Moreover, our picture not only explains the meaning of $\mu_0$, it also demonstrates the main difference between the two impact regimes: the inviscid regime is stabilized by the surface tension and the viscous regime is stabilized by the liquid viscosity.

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FIG. 5. (Color online) The ratio between the destabilizing and the stabilizing stresses, $\Sigma_G/\Sigma_\mu$, measured at $t = t_{on}$, for various pressures and viscosities. All experiments are done under almost identical impact conditions, with only the pressure being varied. Different symbols represent liquids of different viscosities: $\mu = 4.65(\bullet)$, $6.7(\bigcirc)$, $9.3(\blacktriangle)$, and $13.2(\times)$ mPa s. The ratio, $\Sigma_G/\Sigma_\mu \sim \rho_G C_0 dl/\mu$, is computed from direct measurements: $\rho_G$ is calculated from $P$, and $d$ is from the best fit to the high-speed images at the time $t_{on}$. Without any fitting parameter, all the ratios are within the narrow range between 3 and 4, confirming that $\Sigma_G$ and $\Sigma_\mu$ are comparable at the time $t_{on}$.

the moment $t_{on}$. This ratio is tested for various pressures and viscosities, as plotted in Fig.5. All experiments are done under almost identical impact conditions, with only the pressure being varied. Different symbols represent liquids of different viscosities: $\mu = 4.65(\bullet)$, $6.7(\bigcirc)$, $9.3(\blacktriangle)$, and $13.2(\times)$ mPa s. For each impact, we obtain $d$ value at $t_{on}$ from the high-speed photography measurements[16]. The air density $\rho_G$ is directly computed from the pressure $P$. The speed of sound in air at room temperature ($20^\circ\mathrm{C}$), $C_G = 343\mathrm{m\ s^{-1}}$, is a constant independent of $P$. Plugging in all the values, we obtain the ratio, $\Sigma_G/\Sigma_\mu$, as plotted in Fig.5. Without any fitting parameter, most data points collapse to the narrow range between 3 and 4. These values prove that $\Sigma_G$ and $\Sigma_\mu$ are indeed comparable at the time $t_{on}$, as our model predicts.

We study the interaction between the air pressure and the liquid viscosity for the impact of a liquid drop on a smooth substrate. For viscous liquids, the impact is stabilized by the viscous stress, $\Sigma_\mu \sim \mu V_e/d$, whose competition with the destabilizing stress determines when the system becomes unstable. By contrast, for inviscid liquids, the stabilizing stress comes from the surface tension, $\Sigma_L \sim \sigma/d$. Interestingly, by inspecting the two different stabilizing stresses, we find that the liquid viscosity plays opposite roles in them. For $\Sigma_L$ in the inviscid regime, we have $\Sigma_L \sim \sigma/d \sim \sigma/\sqrt{\pi t} \propto 1/\sqrt{\mu}$. Here larger $\mu$ leads to larger $d$ and smaller $\Sigma_L$, thus more viscous liquids are less stable. However, in the viscous regime, we have $\Sigma_\mu \sim \mu V_e/d \propto \sqrt{\mu}$[17]. Now increasing $\mu$ will increase $\Sigma_\mu$ and make the system more stable. This non-monotonic behavior was already observed by previous experiments(see ref. [9] Fig.5) and now can be fully understood.

In summary, our study shows that the interplay between air and liquid viscosity is crucial in determining the outcomes of liquid-solid impacts. The viscosity plays different roles in different regimes, and the simple intuition that a more viscous liquid is more stable during an impact is not always valid.

We gratefully acknowledge helpful discussions with Sidney Nagel, Wendy Zhang, Michelle Driscoll, Alexis Berges and Emily Ching. This project is supported by RGC Research Grant Direct Allocation (Project Code: 2060395), MRSEC DMR-0213745 and NSF DMR-0352777.

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[15] We note that $\Sigma_\mu \propto 1/t$ while $\Sigma_L \propto 1/d \propto 1/\sqrt{\mu}$. Thus $\Sigma_\mu$ always dominates for small $t$. However, $\Sigma_\mu$ decreases more rapidly with $t$ and can drop below $\Sigma_L$ soon after the impact, for the case of small $\mu$. Therefore, $\Sigma_L$ can be dominant for most period of the impact in the inviscid regime.
[16] Due to the discreteness of the measured $d$ values, we use the calculated $d$ value at $t = t_{on}$ from the best fitting function such as the solid line shown in Fig.3 lower inset.
[17] Actually $V_e$ also depends on $\mu$. But the dependence is much weaker than $\sqrt{\mu}$ (unpublished data), and qualitatively our argument is not affected. We also note that in Eq. 1, $V_e$ cancels out, thus its dependence on $\mu$ does not affect the derivation of Eq. 2.