Droplet rebound from rigid or strongly hydrophobic surfaces

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Abstract. – A weakly deformable droplet impinging on a rigid surface rebounds if the surface is intrinsically hydrophobic or if the gas film trapped underneath the droplet is able to keep the interfaces from touching. A simple, physically motivated model inspired by analysis of droplets colliding with deformable interfaces is proposed in order to investigate the dynamics of the rebound process and the effects of gravity. The analysis yields estimates of the bounce time that are in very good quantitative agreement with recent experimental data (Okumura et. al., (2003)) and provides significant improvement over simple scaling results.

Introduction. – The collision and rebound of droplets has been a subject of interest and investigation for many decades. Impacting drops undergo energetic bounces if the pressure in the gas film separating the drops deforms the drop surfaces sufficiently to transform the drop’s kinetic energy into deformation energy before contact occurs. Collisions in an incompressible, continuum gas usually result in a bounce with contact occurring due to inter-particle forces or surface imperfections\textsuperscript{1,2}. Bouncing can also result when drops impinge on rigid super hydrophobic\textsuperscript{3} surfaces or on rigid walls heated to well above the Leidenfrost temperature\textsuperscript{4,5}. Very recently, Okumura et. al.\textsuperscript{3}, presented experimental data for the collision and rebound, in air, of 400 – 1000 µm water drops from a super-hydrophobic surface. They found that the measured bounce time was consistently larger than that predicted by simple scaling. Furthermore, with the drop radius held fixed, the contact time was seen to increase significantly as the velocity of impact decreased. This was attributed to effects of finite drop size and gravity.

In this letter, a physically motivated model is proposed to investigate this scenario, specifically the dependence of the bounce time and deformation on droplet size and velocity. Use of the methodology developed in similar problems involving droplets colliding with deformable interfaces\textsuperscript{1,2}, indicates that even when gravity is absent the bounce time for a liquid drop increases as the impact velocity decreases. Consideration of gravity induced effects suggests that the bounce time is modestly modified relative to the zero-gravity value. The predicted results are in excellent quantitative agreement with experimental data of Okumura et. al.\textsuperscript{3} showing that both effects are needed to account properly for the details of the bounce.
Dimensionless analysis yields reduced parameter space. – Consider a droplet of radius \( a \) comprised of liquid with density \( \rho_d \), viscosity \( \mu_d \) and interfacial tension \( \sigma \) colliding with a rigid flat surface at impact speed \( U_c \). Let the ambient gas be incompressible and continuum with density \( \rho_g \), pressure \( p_o \) and viscosity \( \mu_g \). Dimensional analysis then indicates that the parameters determining the characteristics of the rebound process are - (a) the Weber numbers based on drop and gas properties, \( W_d \equiv \rho_d U_c^2 a \sigma \) and \( W_g \equiv \rho_g U_c^2 a / \sigma \) that measure the relative importance of inertial pressures and surface pressure, (b) the capillary numbers based on drop and gas viscosities, \( C_d \equiv \mu_d U_c / \sigma \) and \( C_g \equiv \mu_g U_c / \sigma \), and (c) the Reynolds numbers \( R_g \equiv a U_c \rho_g / \mu_g \) and \( R_d \equiv a U_c \rho_d / \mu_d \) characterizing viscous effects in the gas and inside the drop. The effects of gravity are quantified by the Froude number, \( F_d \equiv U_c^2 / (ag) \). Droplet inertia is quantified by the Stokes number \( S \sim O(W_d/C_g) \), that also measures the effectiveness of gas viscosity in dissipating the kinetic energy of the drop.

A large number of physically relevant collisions occur at conditions satisfying \( W_d \ll 1 \), \( S \gg 1 \), \( R_d \rightarrow 0 \), \( R_g \leq 1 \), \( W_d^{1/2} / R_d \rightarrow 0 \) and \( C_g \ll 1 \). In such cases, viscous dissipation inside the drop and in the gas outside may be neglected during the collision process as long as the droplet rebounds\(^1\). That is, the dissipation in the gas film has a small impact on the bounce provided we are far from the bounce-contact transition. The primary role of the film is then to cushion the droplet from physical contact with the surface. We expect the dynamics to primarily depend on just two parameters - \( W_d \) and \( F_d \) and seek a theoretical description of the approach and rebound in this special limit.

Collisions in zero gravity. – Consider for now collisions in gravity-less conditions so that \( F_d = \infty \). It is useful to first visualize the approach and rebound scenario - for simplicity, let us choose a collision occurring in the presence of a gas. For \( S \gg 1 \), changes in the droplet velocity occur only when the gap thickness, \( h_o \ll a \), as a result of the large lubrication force in the gap. Drop deformation becomes important when the gap thickness is \( O(aC_g^{1/2}) \ll a \). This is easily seen by balancing the viscous force exerted by the gas with the capillary pressure \( O(2\sigma / a) \). The center of mass speed is chosen to be \( U_c \) at this point. As the gap becomes smaller, the confined air film leads to a strong localized pressure that is \( O(2\sigma / a) \), that when seen from far seems to act over a temporally varying nearly flat region of radial extent \( r_d(t) \) as depicted in Figure (1a). Closer examination of the near-contact region as depicted in Figure (1b) reveals that the inner region may in fact have a more complicated shape - for instance during the
approach, a dimple like shape is seen with a radius of roughly \( r_d(t) \) and a gap thickness that is a maximum at the centerline. Insofar as the bounce process itself is concerned, the detailed shape of the inner region is of secondary importance - what is important is the angular extent of the flat region\(^1\). As the radius of the flat region increases, the center of mass moves slower as as more of the initial kinetic energy is channeled into surface deformation modes. At some point, the centers of mass of the drops reverse their direction of motion and the drops begin to rebound - during this process the flat region begins to shrink and finally vanishes. For a droplet colliding with a super-hydrophobic surface we expect a similar picture to apply. The reason being that due to the nature of the surface-drop interaction, the contact angle is nearly \( 180^\circ \) and the near-contact area is flat to leading order here as well. Thus, drop is nearly spherical except at a small region where it is flat. This shape is reminiscent of the shape found by Mahadevan and Pomeau\(^8\) for small drops rolling due to gravity.

Simple scalings\(^1,2,3\) for the bounce time and deformation are readily obtained. For the deformation energy to balance the \( O(2\pi \rho_d a^3 U_c^2 / 3) \) initial kinetic energy, the characteristic magnitude of the interface deformation should scale as \( O(\rho_d U_c^2 a^3 / \sigma)^{1/2} \). The time scale for the deformation to relax is approximately \( O(\rho_d U_c^2 a^3 / \sigma)^{1/2} \). This value, which we denote by \( \tau_B \), was first obtained by Lord Rayleigh in his study of the vibrational modes of a drop\(^7\). Note that the Rayleigh time scale is independent of the impact velocity. The angular extent of the flat region is obtained by a force balance on the drop and is \( O(\rho_d U_c^2 a^3 / \sigma)^{1/4} \).

To obtain a more accurate picture of the rebound process, the equations for the flow inside the drop have to be solved. The formulation follows directly from earlier work on collisions with deformable interfaces\(^1,2\). A spherical coordinate system that is fixed in space and time in the interior of the droplet, such that the origin lies at a distance \( a \) from the rigid surface, is chosen. The velocity field in the drop is divergence free and governed by the Navier-Stokes equation. Scaling all lengths by \( a \), time with \( a W e_d^{1/2} U_c^{-1} \), and the pressure by \( (\rho_d U_c^2)^{1/2} \) and recognizing that the \( O(a^2 \rho_d U_c^{-1}) \) time scale for viscous diffusion of momentum in the drop is much larger than the bounce time, we find that the fluid in the drop undergoes inviscid impulsive motion described by,

\[
\partial_t \mathbf{u}_d = -\nabla p_d. \tag{1}
\]

Invoking the concept of a velocity potential, \( \phi \) such that \( \mathbf{u}_d = \nabla \phi \), we find that \( \nabla^2 \phi = 0 \) and the dimensional pressure field is given by

\[
p_d = p'_o + 2(\sigma / a) - \rho_d \partial_t \phi',
\]

\( p'_o \) being the ambient pressure. The effect of the \( O(2\sigma / a) \) pressure field localized in a flat region of angular extent \( \alpha(t) \) is incorporated via the equation \( p'_o = p'_o + 2(\sigma / a)H(\alpha(t) - \theta) \), \( \theta \) being the angular co-ordinate of a cylindrical co-ordinate system as in Figure (1a), and \( H \) being the Heaviside function. The radial position vector of a surface point at \( \eta = \cos \theta \) and the velocity potential are expanded using the Legendre functions \( P_k \) of order \( k \) and are given by

\[
r^*_s(\eta, t) = a R(\eta, t) = a(1 + W_d^{1/2} \sum_{k=0}^{\infty} D_k(t) P_k(\eta)) \quad \text{and} \quad \phi = \sum_{k=0}^{\infty} B_k(t)(r^*)^k P_k. \tag{2}
\]

Application of the kinematic and normal stress boundary conditions, the constant volume constraint along with simple geometry then yields

\[
d_t(B_k) = -[(P_{k-1}(\cos \alpha) - P_{k+1}(\cos \alpha))] W_d^{-1/2} - [k(k + 1) - 2] D_k, \quad d_t(D_k) = kB_k,
\]
Weber number, $W_d$ - - - - - -
Scaled bounce time, $T_b = T'_b/\tau_R$

Fig. 2 – Scaled rebound time, $T_b = T'_b/\tau_R$ as a function of Weber number, $W_d$. The solid curve is
the theoretical prediction for $S \to \infty$, $W_d \ll 1$ and $F_d = \infty$. The symbols are experimental data
points$^3$ for water drops in air. The squares correspond to $a = 0.6$ mm, the stars to $a = 0.4$ mm and
the circles to $a = 1$ mm.

Equation set (3) is then solved in conjunction with the initial conditions $D_{k \geq 1}(t = 0) = 0$,
$B_{k \geq 2}(t = 0) = 0$ and $B_1(t = 0) = 1$. The rebound is complete when the center of mass
of the droplet reaches its initial position viz., $D_1(t = T_b) = 0$. Numerical solutions are obtained
for various values of $W_d$ by using a second-order predictor-corrector method and varying the
number of modes until convergent results are obtained.

Figure (2) summarizes the results for $F_d = \infty$ and compares them to experimental obser-
vations of Okumura et.al.$^3$. The solid line corresponds to the theoretical prediction obtained
by numerically solving equation (3). We notice immediately that in spite of the different
impact velocities and sizes, there is a reasonable collapse of the experimental data around the
theoretical prediction, indicating that $W_d$ is the relevant parameter to use. Good qualitative
and quantitative agreement is seen, especially for the smaller size drops. Some of the data
points for the 1 mm drop do differ considerably from the predicted values but the reasons for
these are not clear. Nonetheless we find that the theoretical result yields a good prediction
even as the Weber number becomes $O(0.1)$.

Both experimental data (that include gravity effects) and the numerical results (without
gravity) consistently indicate that as $W_d$ decreases, the bounce time increases with
decreasing $U_c$. To investigate this behavior in more detail, analytical asymptotes for the
bounce time were obtained by a singular perturbation analysis of (3). It is found that when
$\ln^{1/2}(W_d^{-1/4}) \gg 1$, all the deformation modes do not scale similarly as $O(aW_d^{1/2})$, instead
a separation of scales develops. The $k = 1$ mode corresponding to translation of the center
of mass scales as $O(aW_d^{1/2} \ln^{1/2}(W_d^{-1/4}))$ while the $k \geq 2$ surface deformation modes scale
as $O(aW_d^{1/2} \ln^{-1/2}(W_d^{-1/4}))$. As a result, when $W_d \to 0$, the highly localized pressure field
causes the $k \geq 2$ flow modes to become negligibly small. The radial extent of the inner region
then couples directly to the $k = 1$ translational mode. In other words, rather than deform uniformly, the approaching drop prefers to flatten the near contact region and shift the center of mass appropriately in response to the ensuing force. A spring like motion manifests itself wherein the center of mass executes a harmonic oscillation but like a non-linear spring that stiffens strongly as $W_d \to 0$. In fact, for asymptotically small $W_d$, the bounce time scales as,

$$T_b' \approx \pi \sqrt{(2/3)} \left[ a U_c^{-1} W_d^{1/2} \ln^{1/2} (W_d^{-1/4}) \right]$$

while the maximum extent of the flat region scales as $\alpha_{\max} = [2W_d/(3 \ln (W_d^{-1/4}))]^{1/4}$. As the Weber number increases, all the deformation modes become equally important and the separation of scales diminishes. The asymptotic expression in (4) consistently under-predicts the actual bounce time since the analytical approximation is obtained by ignoring the $k = 2$ modes. For finite $W_d$, the symmetry between approach and rebound is broken. The effects of finite Weber number render the collision process asymmetric, increasingly so as the Weber number approaches $O(1)$ values.

First effects of gravity. – The effects of gravity cannot be ignored when $W_d \sim O(F_d^2)$ i.e., when the impact velocity is $O(a^2 g^2/\sigma)^{1/2}$ or smaller. To anticipate the effects of gravity we consider the case of weak but non-zero gravity. That is, $F_d$ is finite but $\varepsilon_1 \equiv W_d^{1/2} F_d^{-1} \ll 1$. Thus the droplet is still weakly deformed and the flow and deformation modes are still described by (2). Note that the small parameter $\varepsilon_1$ is the ratio of the critical speed at which gravity becomes important and the impact speed - that is $\varepsilon_1 \equiv ag(\rho g/\sigma)^{1/2} U_c^{-1} \equiv V_g/U_c$. A measure of the importance of gravity in affecting the bounce time is then obtained by scaling the actual bounce time, $T_b'$ with the gravity-free value $T_b^0$ and then plotting this normalized rebound time as a function of a suitable small parameter. Since the data for 400 micron drops correspond most closely to the limits we are investigating, these are chosen to test the theoretical predictions.

Okumura et. al., proposed an approximate expression to incorporate the effects of gravity which may be written as

$$\left( \frac{T_b'}{T_b^0} \right)_o = 2 \left( 1 - \frac{1}{\pi} \cos^{-1} \left( \frac{\varepsilon_1}{(1 + \varepsilon_1^{2})^{1/2}} \right) \right).$$

In their expression, the zero-gravity bounce time, $(T_b^0) = (2.3 \tau_R)$ is independent of $W_d$ and thus does not depend on the impact velocity. The bounce time as predicted increases by at most a factor of 2 compared to the zero-gravity value. This expression is plotted as a dashed curve in figure (3a) and compared to the experimental obtained data points normalized the same way. The circles indeed map onto to small $\varepsilon_1$ values but the normalized bounce times are all much greater than unity. At first sight this may seem to imply that gravity plays a crucial role - however this is not quite correct. Since the true zero-gravity bounce time is dependent on $W_d$, a more accurate reflection of gravity effects is obtained upon incorporating this dependence.

Figure 3b illustrates precisely this effect. The squares are the re-normalized data points obtained by calculating $T_b^0$ corresponding to the solid line in figure 2, and plotting $T_b'/T_b^0$ as a function of the small parameter $\varepsilon_2 \equiv W_d^{1/2} \ln^{1/2} (W_d^{-1/4}) F_d^{-1}$. The data points map to low values of $\varepsilon_2$ as expected indicating the weak effects of gravity. More importantly, the scaled bounce times are within 20 % of the gravity-free value suggesting that the quantitative effects of gravity are relatively modest.

An approximate theoretical treatment allows us to anticipate the dependence of the scaled bounce time on $\varepsilon_2$ observed in figure 3b. For asymptotically small Weber number, the center
Fig. 3 – The effects of gravity on the rebound time are explored in this plot. (a) Comparison of equation (5) proposed by Okumura et. al. (dashed curve) with the normalized bounce times (open circles) for 400 micron droplets colliding in air. The x axis is \( \varepsilon_1 \equiv V_g U_c^{-1} \). The gravity-free bounce time is independent of \( W_d \). (b) The solid line corresponds to equation (8) and incorporates the effect of \( W_d \) on the gravity-free bounce time. The squares are the same data points re-normalized differently and plotted as a function of \( \varepsilon_2 \equiv V_g \ln^{1/2}(W_d^{-1/4})U_c^{-1} \).

of mass of the drop behaves as if attached to a spring with a \( W_d \) dependent stiffness. When the effects of an external gravity field are included, an additional force acts on the center of mass due to the weight of the drop. Set \( r_s(\eta, t) = a(1 + \delta D_1 P_k(\eta) + O(\delta_2)) \) where \( \delta_1 \equiv W_1 d \ln^{1/2} (W - 1/4) d U_c^{-1} \) and \( \delta_2 \equiv W_1 d \ln^{-1/2} (W^{-1/4}) \). The force balance on the center of mass of the drop has the form

\[
(4 \pi \rho_d a^3 / 3)(a \delta_1 [U_c^{-1}(a \delta_1)]^{-2} d_1^2(D_1) = -2(\sigma / a) \pi (a \delta_2 / 2)^2 a^2 + (4 \pi \rho_d a^3 g / 3)(g \cdot U_c) (g U_c)^{-1}.
\]

Algebraic manipulations using asymptotic properties of the Legendre polynomials show that \( \alpha^2(t) \approx D_1(t) + O(\delta_2) \). The deformation mode \( D_1 \) then satisfies

\[
d_1^2(D_1) = -\frac{3}{2} D_1 + \frac{\delta_1}{F_d} (g \cdot U_c)(g U_c)^{-1} = -\frac{3}{2} D_1 + \varepsilon_2
\]

the small parameter \( \varepsilon_2 \equiv \delta_1 / F_d \) and gravity has been chosen to act in the direction of the initial droplet motion. Equation (5) indicates that the relevant small parameter quantifying effects of gravity when \( W_d \ll 1 \) is not \( \varepsilon_1 \) but \( \varepsilon_2 \). Solving (5) with the initial conditions \( D_1(t = 0) = 0 \) and \( (d_1 D_1)(t = 0) = 1 \) yields the solution

\[
D_1(t) = \frac{2}{3} \varepsilon_2 + \frac{2}{3} \left( 1 + \frac{2}{3} \varepsilon_2^2 \right) \frac{1}{2} \sin \left( \sqrt{\frac{3}{2}} t \right) + \tan^{-1} \left( -\varepsilon_2 \sqrt{\frac{2}{3}} \right) \right).
\]

This corresponds to the drop being spherical and moving with speed \( U_c \) when the collision process starts.

It is easy to show from equation (6) that when \( 0 < \varepsilon_2 \ll 1 \), the modified bounce time is given by

\[
\frac{T_b'}{T_b^0} = \left( 1 - \frac{2}{\pi} \tan^{-1} \left( -\varepsilon_2 \sqrt{\frac{2}{3}} \right) \right) \]

It is clear that the bounce time increases relative to the zero-gravity value by a factor that is between 1 and 2, the latter value corresponding to \( \varepsilon_2 \to \infty \).
Equation (8) shows surprisingly good agreement with the experimental data points which seems to suggest that gravity acts as a weak perturbation for these droplets. Perhaps, a more careful calculation in lieu of the simplified treatment just described will result in a better prediction for larger values of $\varepsilon$. Also note that the dashed and solid curves seem to have the same form - this is of course primarily due to the way we have chosen to normalize the curves. It should also be noted that if the direction in which gravity acts is not the same as that of the initial droplet motion, axi-symmetry is broken and the bounce times can differ noticeably.

Summary and conclusions. – To summarize, the collision and rebound dynamics of weakly deforming drops impacting with velocities, $U_c$, satisfying $(a^3\rho g^2/\sigma) \ll U_c^2 \ll (a\rho/\sigma)^{-1}$, is controlled by the interplay between the kinetic energy of the drop and energy stored in surface deformation due to finite surface tension, $\sigma$ with gravity acting as a weak perturbation. In the limit of asymptotically small $W_d$, the scaled bounce time $T_b = T_b(\rho a^3/\sigma)^{-1/2}$, diverges as $\ln^{1/2}(\rho U_c^2 a/\sigma)^{-1/4}$ when $U_c \to 0$. To leading order, gravity tends to increase the time scale for the bounce relative to the zero-gravity value. Results of numerical calculations indicate that the bounce time increases relative to the asymptote as $U_c^2 \to (a\rho/\sigma)^{-1}$ - a manifestation of the increasing effect of non-linear deformation modes and of the drop being deformed everywhere rather than merely locally.

Is the $O(aW_d^{1/4})$ local flattening seen in the $W_d \ll 1$ case, a precursor of the global deformation seen in pancake shaped drops for $W_d \geq 1$? It is tempting to think that as $W_d \to 1$, the maximum extent of the inner region grows till it becomes comparable to the drop radius $a$ with the transition between the inner and outer region becoming gentler. Consider increasing the impact speed, $U_c$ such that it becomes $O(a\rho/\sigma)^{-1/2}$ as happens when $W_d \sim O(1)$. In this limit, gravity plays a negligible role. The strongly deforming droplet changes to a pancake like shape with characteristic radius $R_d \geq a$ being different from the height $H_d \sim O(a^3R_d^{-2})$. The initial kinetic energy is transferred mainly into internal flow modes within the drop. The maximum extent of the pancake drop is determined by a balance between the flow driven by inertial acceleration and the flow due to the surface tension induced pressure gradient at the edge of the pancake. This yields the balance $(\rho aU_c^2/a) \sim (\sigma R_d^4/a^6)$ thus yielding $(R_d/a)^4 \sim O(\rho aU_c^2 a/\sigma) \sim O(W_d)$. Although the scalings for the maximal apparent contact area seem to arise differently for cases $W_d \ll 1$ and $W_d \gg 1$, it is interesting and in some sense satisfying to note that they exhibit the same dependence on $W_d$.

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REFERENCES

[1] Gopinath A and Koch D. L., J. Fluid Mech., 454 (2002) 145.
[2] Gopinath A and Koch D. L., Phys. Fluids, 13 (2001) 3526.
[3] Okumura K., Chevy F., Richard D., Quere D. and Clanet C., Europhys. Lett., 62 (2003) 237.
[4] Karl A. and Frohn A., Phys. Fluids, 12 (2000) 785.
[5] Yao S.-C. and Cai K. Y., Exp. Therm. Fluid Sci., 1 (1988) 363.
[6] Rayleigh, Lord, Proc. R. Soc. Lond. A., 29 (1879) 71.
[7] Mahadevan L. and Pomeau Y., Phys. Fluids, 11 (1999) 2449.