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Droplet detachment force and its relation to Young-Dupre adhesion

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Abstract

Droplets adhere to surfaces due to their surface tension $\gamma$ and understanding the vertical force $F_d$ required to detach the droplet is key to many technologies (e.g., inkjet printing, optimal paint formulations). Here, we predicted $F_d$ on different surfaces by numerically solving the Young-Laplace equation. Our numerical results are consistent with previously reported results for a wide range of experimental conditions: droplets subjected to surface vs. body forces with $|F_d|$ ranging from nano- to milli-newtons, droplet radii $R$ ranging from tens of microns to several millimetres, and for various surfaces (micro-/nano-structured superhydrophobic vs. lubricated surfaces). Finally, we derive an analytic solution for $F_d$ on highly hydrophobic surfaces and further show that for receding contact angle $\theta_r > 120^\circ$, the normalized $F_d/\pi R$ is equivalent to the Young-Dupre work of adhesion $\gamma(1 + \cos \theta_r)$.

Introduction

We encounter the effects of droplet adhesion every day: dew of droplets can be seen clinging to blades of grass in early morning, and droplets stuck to camera lenses affect image clarity on rainy/misty days. 3,12 Despite the prevalence of the phenomenon and clear importance in various technologies (from inkjet printing3 to agricultural technologies4,5), there is still no consensus on how to link the vertical force $F_d$ required to detach a droplet from a surface to its wetting properties. 6–9 For example, Tadmor et al. (2017) proposed that $F_d/2\pi r$ (where $r$ is the contact radius) is equivalent to the Young-Dupre work of adhesion $\gamma(1 + \cos \theta_r)$ where $\gamma$ is the surface tension and $\theta_r$ is the receding contact angle,10 but others disagreed.11–13

Figure 1: Different methods to measure droplet detachment force $F_d$ using (A) atomic force microscope (volume $V = 65 \mu$L, $U = 2 \mu$m s$^{-1}$, superhydrophobic surface), (B) microbalance force sensor ($V = 5 \mu$L, $U = 5 \mu$m s$^{-1}$, superhydrophobic surface), and (C) centrifugal adhesion balance ($V = 10 \mu$L, hydrophobic). A is original unpublished data, while B and C are taken from Daniel et al. (2023)7 and Tadmor et al. (2017),10 respectively.

Extensive experimental data for droplet de-
attachment exist in the literature, but their interpretation is complicated by the fact that various groups use very different methods to measure $F_d$ (Fig. 1) with very different droplet volumes ($V = 5\ \text{pL} - 10\ \mu\text{L}$) and detachment speeds ($U = 10^{-6} - 10^{-3}\ \text{m s}^{-1}$). As a result, different groups reported force magnitudes that vary considerably from 38 nN (as measured using atomic force microscopy or AFM) to 1.8 $\mu$N (force microbalance), and 0.36 mN (centrifugal adhesion balance or CAB). The various approaches described in the literature can be broadly categorized into surface vs. body force methods—an important distinction that is often ignored. Methods such as AFM and microbalance force sensor (Fig. 1A, B) apply a pulling force only at the top surface of the droplet, i.e., surface force. In contrast, in CAB (and other methods), the centrifugal pulling force is applied to the entire droplet, i.e., body force.

In this paper, we show that the quasi-static approximation applies during the retraction process, and that $F_d$ can be predicted by numerically solving the Young-Laplace equation. Our numerical results are consistent with the experimental data collected independently by various research groups. We derive an analytic solution for $F_d$ and further show that for a droplet of radius $R$ the normalized $F_d/\pi R$ (as opposed to $F_d/2\pi r$) is equivalent to the Young-Dupre work of adhesion $\gamma(1 + \cos \theta_r)$ for highly hydrophobic surfaces with $\theta_r > 120^\circ$. In contrast, for less hydrophobic surfaces with $\theta_r < 120^\circ$ there is no simple way to relate $F_d$ to the Young-Dupre work of adhesion.

Our analysis is general and applies for a wide range of experimental conditions (including those in Figure 1) and different surfaces (flat, micro-/nano-structured superhydrophobic, and lubricated surfaces). We have described the method at length in our previous publications. Please see our previous publications.

Methods

Surface preparation.

To make the superhydrophobic surface in Fig. 5A, we started with a 3 mm thick Sylgard 184 polydimethylsiloxane (PDMS) slab, spraycoated it with a layer of hydrophobic nanoparticles (Glaco Mirror Coat Zero, Soft 99 Co.) before placing it vertically to dry for one hour before use. To make the lubricated surface in Fig. 5B, we coated a thin layer of fluorinated oil (GPL Krytox 101, ~3 $\mu$m thick) onto the nanostructured PDMS slab.

Measuring detachment force.

To measure the detachment force using AFM, please see our previous publications. We also measure detachment force due to gravity by first puncturing the PDMS slab with a 34 Gauge needle (outer diameter of 0.16 mm), placing the surface upside down, and using a syringe pump to slowly increase the droplet volume until the droplet detaches (due to gravity).

Numerical simulations.

To solve the axi-symmetric Young-Laplace equation, we use the shooting method implemented in the Python programming language. We have described the method at length in our previous publication. Python codes used here are provided in github repositories.

Results and discussions

We first find $F_d$ by numerically solving the axisymmetric Young-Laplace equation for a droplet subjected to either a surface force (Fig. 2) or a body force (Fig. 3). We can recast the Young-Laplace equation into its non-dimensional form by normalizing the various quantities with $V^{1/3}$ and surface tension $\gamma$ (e.g., $\tilde{u} = u/V^{1/3}$, $\Delta \tilde{P} = \Delta PV^{1/3}/\gamma$) to give

$$\frac{\tilde{u}''}{(1 + \tilde{u}'^2)^{3/2}} - \frac{1}{\tilde{u}' \sqrt{1 + \tilde{u}'^2}} = -\Delta \tilde{P} \text{ for } \tilde{z} \in (0, \tilde{h}),$$

$$\tilde{u}(\tilde{h}) = \tilde{r},$$

$$\tilde{u}'(\tilde{h}) = -\cot \theta_r,$$

$$\int_0^{\tilde{h}} \pi \tilde{u}'^2 d\tilde{z} = 1$$

(1)
Figure 2: (A) Schematic of a droplet attached to a disc of radius $a$ and retracting from the surface. (B) Numerical solutions to Young-Laplace equation showing the non-dimensional force $F/\gamma V^{1/3}$ as a function of increasing $h/V^{1/3}$ for different contact angles $\theta_r = 40^\circ$–$160^\circ$. Inset shows the magnified plot for $\theta_r = 160^\circ$. (C)–(E) Droplet geometries at the point of detachment for different $\theta_r$. Scale bar in C is the non-dimensional capillary length $\sqrt{\bar{F}_d} = 2.64$ for $\theta_r = 2^\circ$.

Figure 3: (A) Schematic of a droplet of volume $V$ and density $\rho$ subjected to a body force $F = V\rho g_{\text{eff}}$ where $g_{\text{eff}}$ is the effective acceleration. (B) Numerical solutions to Young-Laplace equation showing the non-dimensional contact radius $r/V^{1/3}$ as a function of increasing $F/\gamma V^{1/3}$ for different contact angles $\theta_r = 2^\circ$–$160^\circ$. Inset shows the magnified plot for $\theta_r = 160^\circ$. (C)–(E) Droplet geometries at the point of detachment for different $\theta_r$. Scale bar in C is the non-dimensional capillary length $\sqrt{\bar{F}_d} = 2.64$ for $\theta_r = 2^\circ$.

where $r$ is the droplet’s contact radius and we assume that the droplet retracts with a constant receding contact angle $\theta_r$.

For the surface force method, we can approximate the droplet geometry as being held by a circular disc with radius $a$ at the top and further impose the boundary condition $\bar{u}(0) = \bar{a}$ (Fig. 2A) and assume that the Laplace pressure $\Delta \bar{P} = \Delta \bar{P}_o$ is constant (i.e., we neglect the effects of gravity since the droplets are typically smaller than the capillary length). We performed numerical simulations for different $\theta_r = 40^\circ$–$160^\circ$ and fixed $\bar{a} = 0.45$ (which approximates well the geometries used in various papers$^{14,15}$ including in Fig. 1A, B) and plotted the force acting on the droplet

$$\tilde{F} = -2\pi \bar{r} \sin \theta + \pi \bar{r}^2 \Delta \bar{P}_o \quad (2)$$

(in its non-dimensional form) as we progressively stretched the droplet (Fig. 2B).

We terminate the simulations when we can no longer achieve numerical convergence; this is the point of droplet detachment and we can define the corresponding detachment force $\bar{F}_d$ which strongly depends on $\theta_r$, i.e., the surface wetting properties. For hydrophilic surfaces, the detachment force is high ($\bar{F}_d = 1.6$ for $\theta_r = 40^\circ$) but can become negligible for superhydrophobic surfaces ($\bar{F}_d = 0.1$ for $\theta_r = 160^\circ$). For hydrophilic surfaces, the droplet is highly
stretched at the point of detachment and resembles more of a capillary bridge (Fig. 2C). In contrast, the droplet retains its spherical cap shape till the point of detachment for highly hydrophobic surfaces (Fig. 2D, E).

Interestingly, the droplet experiences a maximum force $\bar{F}_{\text{max}} > \bar{F}_d$ before detaching. For hydrophilic surfaces, the two force quantities can be quite different, with $\bar{F}_{\text{max}}/\bar{F}_d = 2.2$ for $\theta_r = 40^\circ$ (Fig. 2B). In contrast, $\bar{F}_{\text{max}}/\bar{F}_d = 1.1$ for $\theta_r = 160^\circ$ (inset in Fig. 2B).

We now repeat the numerical simulations for a droplet subjected to an effective acceleration $g_{\text{eff}}$ and an equivalent non-dimensional body force

$$\bar{F} = V^{2/3} \rho g_{\text{eff}}/\gamma$$  \hspace{1cm} (3)

The droplet geometry still obeys the Young-Laplace equation in Equation 1, except with different boundary conditions $\bar{u}(0) = \bar{u}'(0) = 0$ and with the Laplace pressure $\Delta P = \Delta P_{\text{d}} - \bar{F} \bar{z}$ that varies with position $\bar{z}$ (Fig. 3A).

We progressively stretch the droplets by increasing $\bar{F}$ and noted the decrease in base radius $\bar{r}$ for different $\theta_r = 2$–$160^\circ$ (Fig. 3B). Note that $\bar{F}$ is the input variable here, whereas it is the output variable in Fig. 2B. As is before, we terminate the simulations when there is no longer numerical convergence, and we can define the corresponding detachment force $\bar{F}_d$ (but no $\bar{F}_{\text{max}}$). For hydrophilic surfaces, the droplet resembles a spherical cap whose base is surrounded by a wetting skirt/meniscus of size $\sqrt{\bar{F}_d}$ the non-dimensional capillary length (See scale bar in Fig. 3C). The wetting skirt is a low pressure region and provides the suction required to hold onto the droplet. In contrast, for highly hydrophobic surfaces, the droplet retains its spherical cap geometry (Fig. 3D, E).

We can check the validity of our simulation results by superimposing experimental data collected independently by five different research groups using different methodologies ranging from AFM,14 microbalance force sensors,15,16 and CAB.10,13 When the normalized detachment force $F_d/\gamma V^{1/3}$ is plotted against $\epsilon = 1 + \cos \theta_r$, all the experimental data are consistent with results from our numerical simulations for both surface and body forces (blue and red curves in Fig. 4A, respectively). We have also included results collected by us (open symbols in Fig. 4A), including previously unpublished results in Fig. 1A and Fig. 5.

Interestingly, the two master curves converge in the limit of $\theta_r > 120^\circ$. In other words, $F_d$ measurement is independent of the method chosen for highly hydrophobic surfaces with $\theta_r > 120^\circ$. In contrast, for less hydrophobic surfaces with $\theta_r < 120^\circ$, $F_d$ measurement values depend strongly on the method chosen (surface vs. body forces), a point that is not well appreciated in the literature.

![Figure 4](image)

Figure 4: (A) Plot of $F_d/\gamma V^{1/3}$ as a function of $(1 + \cos \theta_r)$. Unfilled data points are our experimental results corresponding to Figures 1A, 5A, 5B and from our previous paper,14 while filled data points are results from various groups.10,13,15,16 (B) Droplet geometry during the detachment process for high $\theta_r$. Raw dataset can be found in.26

For convenience, the numerical solutions of $F_d$ for surface and body force methods can be
fitted with polynomial functions

\[
\frac{F_d}{\gamma V^{1/3}} = \begin{cases} 
1.971\epsilon - 0.961\epsilon^2 + 0.190\epsilon^3 & \text{surface} \\
2.296\epsilon - 0.791\epsilon^2 + 0.696\epsilon^3 & \text{body} 
\end{cases}
\]

Relation of \( F_d \) to Young-Dupre work of adhesion

We can derive an analytic expression for \( F_d \) in terms of \( \epsilon = 1 + \cos \theta_r \) for \( \epsilon \ll 1 \), i.e., in the limit of \( \theta_r \to 180^\circ \). According to Young-Dupre, the energy required to detach the droplet is given by \( \Delta E_\gamma = \pi R^2 \gamma \epsilon \), which assumes that the droplet retains its spherical cap geometry after detachment. This is a reasonable assumption for a frozen water droplet detaching from a cold surface, but not for liquid droplets at room temperature. In reality, the detached droplet adopts a spherical shape (Fig. 4B); using simple geometrical arguments and power series expression, we can show that \( \Delta E_\gamma \approx \pi R^2 \gamma \epsilon^2 \). At the same time, the droplet’s centroid is raised by an amount \( \delta z = z' - z \approx R \epsilon \) (See full derivation in Supporting Figure S1).

Since the work done by the detachment force \( F_d \delta z \) must be equal to \( \Delta E_\gamma \),

\[
F_d = \pi R \gamma \epsilon \frac{F_d}{\gamma V^{1/3}} = \left( \frac{\pi}{2} \right)^{2/3} 3^{1/3} \epsilon^{2/3} \approx 1.95 \epsilon 
\]

using the fact that \( V = 4/3\pi R^3 \) (Compare Equations 5 and 4). The analytic expression in Equation 5 (gray curve in Fig. 4) agrees well with the experimental data and the two numerical solutions to Young-Laplace equation for \( \epsilon < 0.5 \) or equivalently \( \theta_r > 120^\circ \). This is not surprising since for high \( \theta_r \), the droplet geometry is well approximated by a sphere (See Figs. 2D, E and Figs. 3D, E).

For highly hydrophobic surface, the detachment force \( F_d \) when normalized by \( \pi R \) is therefore equivalent to \( \gamma (1 + \cos \theta_r) \) the Young-Dupre work of adhesion (per unit area). This result is different from the scaling proposed by Tadmor et al. (2017) where the authors suggested that \( F_d/2\pi r = \gamma (1 + \cos \theta_r) \). For hydrophilic surfaces, there is no simple way to relate \( F_d \) to \( \cos \theta_r \) since the droplet is highly deformed and its geometry depends on the choice of experimental method (compare Fig. 2C and Fig. 3C).

Figure 5: Droplet detachment due to gravity. Water droplets at the point of detachment for (A) superhydrophobic and (B) lubricated surfaces. Raw dataset can be found in.

The results presented here are general and apply for a wide range of experimental conditions and surface types from superhydrophobic (Fig. 5A) to lubricated surfaces (Fig. 5B). In Fig. 5, we slowly increase the droplet volume until gravity (another example of body force) causes the droplet to detach from the surface.

Validity of quasi-static approximation

In our analysis so far, we have used the quasi-static approximation and assumed that the body force applied or the gap \( h \) is increased gradually such that any dynamic effects can be ignored. \( F_d \) is dominated by surface wetting properties (in particular \( \theta_r \)) and viscous dissipation is negligible. This is a reasonable assumption in most cases and explains why different groups reported that \( F_d \) is independent of \( U \) for superhydrophobic surfaces.\(^{14,15}\)

There are however cases where this assumption is not valid. For example, some underwater superoleophobic surface exhibits \( \theta_r = 180^\circ \) (i.e., no contact line pinning) and \( F_d \) is always dominated by viscous dissipation. Previously,
we showed that $F_d \propto U^{3/5}$ for an oil droplet detaching from such a surface.\textsuperscript{23,28}

Maximum vs. detachment force

![Figure 6: Comparison between numerical results for $\tilde{F}_{\text{max}}$ (dashed blue line) and experimental results from Samuel et al. (2011)\textsuperscript{16} and Zhu et al. (2022).\textsuperscript{17} Results in Fig. 4 are also superimposed (in faint) for easy comparison.](image)

As discussed previously, for surface force method (but not for body force), the droplet experiences a maximum force $F_{\text{max}} > F_d$. There is much less data available for $F_{\text{max}}$ in the literature. Nevertheless, the experimental results from Samuel et al. (2011)\textsuperscript{16} and Zhu et al. (2022)\textsuperscript{17} are consistent with our numerical results (Fig. 6).

Our numerical simulations indicate that $F_{\text{max}}$ is linearly proportional to $\epsilon$, i.e.,

$$\frac{F_{\text{max}}}{\gamma V^{1/3}} = 1.971 \epsilon$$

(dashed blue line, Fig. 6). Our numerical results therefore suggest that $F_{\text{max}}$ (unlike $F_d$) can be related to $\gamma(1 + \cos \theta_r)$ even for hydrophilic surfaces, though more experimental data is required to confirm this.

Detachment vs. friction force

It is important to recognize that the lateral $F_{\text{fric}}$ acting on a droplet can be very different in magnitude from the vertical $F_d$ discussed in this paper. Furmidge proposed that the lateral friction force acting on the droplet is given by the relation

$$F_{\text{fric}} = \pi r \gamma (\cos \theta_r - \cos \theta_a)$$

(7)

where $\theta_a$ is the advancing contact angle. For superhydrophobic surfaces which have $\theta_a \approx 180^\circ$, $F_{\text{fric}} \approx \pi r \gamma (1 + \cos \theta_r)$.\textsuperscript{29} Hence, $F_d/F_{\text{fric}} = R/r \sim 10$, i.e., it is easier to move a droplet on a superhydrophobic surface by applying a lateral force as compared to a vertical force.

More recently developed surfaces, such as lubricated\textsuperscript{21,22} and slippery covalently-attached liquid (SCAL) surfaces,\textsuperscript{30} typically exhibit $\theta_r \sim 90^\circ$, $r \sim R$, and $1 + \cos \theta_r \sim 1$, $\cos \theta_r - \cos \theta_a \sim 10^{-3}$.\textsuperscript{31–33} Hence, $F_d/F_{\text{fric}} \sim 10^3$.

Conclusions

Using a combination of numerical simulations and simple geometrical arguments, we were able to explain the observed detachment forces reported by various research groups spanning over a wide range of experimental parameters from micron- to millimetric-sized droplets and force magnitudes ranging from nano- to millinewtons. We also showed that for high hydrophobic surfaces with $\theta_r$, the normalized detachment force $F_d/\pi R$ is equivalent to the Young-Dupre adhesion $\gamma(1 + \cos \theta_r)$.

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Supporting information for
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Analytic solution for detachment force

Here, we will derive an expression for the non-dimensional detachment force \( \tilde{F}_d = F_d/\gamma V^{1/3} \) as a power series of \( \epsilon = 1 + \cos \theta_r \). Our analytic solution applies for droplets with high contact angles, i.e., where \( \epsilon \ll 1 \).

At the onset of detachment, the droplet geometry can be modelled as a spherical cap of radius \( R \), with the volume \( V \) and centroid position \( z \) given by

\[
V = \frac{\pi}{3} R^3 (2 + \cos \theta_r)(1 - \cos \theta_r)^2
= \frac{\pi}{3} R^3 (1 + \epsilon)(2 - \epsilon)^2
\]

(S1)
FIG. S1: Droplet detachment in a) Young-Dupre Model and b) our model.

and

$$z = \frac{3R(1 + \cos \theta_r)^2}{4(2 + \cos \theta_r)} - R \cos \theta_r$$
$$= \frac{3R\epsilon^2}{4(1 + \epsilon)} - R(\epsilon - 1)$$
$$= R \left(1 - \epsilon + \frac{3\epsilon^2}{4(1 + \epsilon)}\right)$$  \hspace{1cm} (S2)

After detaching, the droplet is now a sphere with radius $R'$ (which also equivalent to the new centroid position $z'$) and by conservation of volume

$$\frac{4}{3}\pi R'^3 = \frac{\pi}{3} R^3 (1 + \epsilon)(2 - \epsilon)^2$$
$$R' = R \left(\frac{(1 + \epsilon)^{1/3}(2 - \epsilon)^{2/3}}{4^{1/3}}\right)$$  \hspace{1cm} (S3)
The droplet’s centroid position has been raised by an amount \( \delta z \)

\[
\delta z = z' - z = R \left( \frac{(1 + \epsilon)^{1/3}(2 - \epsilon)^{2/3}}{4^{1/3}} - \frac{3 \epsilon^2}{4(1 + \epsilon)} \right) - R \left( \epsilon - \epsilon^2 - \frac{2 \epsilon^3}{3} + O(\epsilon^4) \right)
\]

There is also an increase in the surface area of the droplet by an amount \( \delta A \), where

\[
\delta A = 4\pi R'^2 - 2\pi R^2(1 - \cos \theta) = 4\pi R^2 \frac{(1 + \epsilon)^{2/3}(2 - \epsilon)^{4/3}}{4^{2/3}} - 2\pi R^2(2 - \epsilon) = 2\pi R^2 \left( \epsilon - \epsilon^2 + \frac{\epsilon^3}{3} + O(\epsilon^4) \right)
\]

Total change in interfacial energy is given by \( \Delta E_\gamma = \pi r^2 (\gamma_s - \gamma_{ls}) + \delta A \gamma \), where \( \gamma \) is the droplet’s surface tension, \( \gamma_s \) is the solid’s surface energy, and \( \gamma_{ls} \) is the liquid-solid surface energy. In the Young-Dupre model, \( \delta A = \pi r^2 \). Here, we use the expression in Equation S5 for \( \delta A \) and the relations \( r = R \sin \theta \), and \( \gamma_s - \gamma_{ls} = \gamma \cos \theta \), to get

\[
\Delta E_\gamma = \pi R^2 \gamma \sin^2 \theta \cos \theta + 2\pi R^2 \gamma \left( \epsilon - \epsilon^2 + \frac{\epsilon^3}{3} + O(\epsilon^4) \right)
\]

\[
= \pi R^2 \gamma (1 - \cos \theta)(1 + \cos \theta) \cos \theta + 2\pi R^2 \gamma \left( \epsilon - \epsilon^2 + \frac{\epsilon^3}{3} + O(\epsilon^4) \right)
\]

\[
= \pi R^2 \gamma (-2\epsilon + 3\epsilon^2 - \epsilon^3) + 2\pi R^2 \gamma \left( \epsilon - \epsilon^2 + \frac{\epsilon^3}{3} + O(\epsilon^4) \right)
\]

\[
= \pi R^2 \gamma \left( \epsilon^2 - \frac{\epsilon^3}{3} + O(\epsilon^4) \right)
\]
detachment force $F_d$ over the distance $\delta z$, i.e.,

$$F_d \delta z = \Delta E_\gamma$$

$$F_d R = \pi R^2 \gamma \left( \epsilon^2 - \frac{\epsilon^3}{3} + O(\epsilon^4) \right) \left( \epsilon - \epsilon^2 - \frac{2\epsilon^3}{3} + O(\epsilon^4) \right)^{-1} \quad \text{(S7)}$$

$$F_d = \pi R \gamma \left( \epsilon + \frac{2\epsilon^2}{3} + O(\epsilon^3) \right)$$

We can recast Equation S1 to get $R = \left(3V/\pi\right)^{1/3}(1 + \epsilon)^{-1/3}(2 - \epsilon)^{-2/3}$ and substituting this to Equation S7 to get

$$F_d = \pi \left( \frac{3V}{\pi} \right)^{1/3} \gamma \left( \frac{\epsilon}{2^{2/3}} + O(\epsilon^2) \right)$$

$$\frac{F_d}{\gamma V^{1/3}} = \left( \frac{\pi}{2} \right)^{2/3} 3^{1/3} \epsilon + O(\epsilon^2) \quad \text{(S8)}$$

$$\tilde{F}_d \approx \left( \frac{\pi}{2} \right)^{2/3} 3^{1/3} (1 + \cos \theta)$$

$$\approx 1.95(1 + \cos \theta)$$

\[1\]
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