Effective interactions between colloidal particles at the surface of a liquid drop

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Pickering Emulsions

- emulsion + colloidal particles
- particles get trapped at the surface of droplets

- applications: stabilization of emulsions, engineering of functional particles
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Stability of a colloidal particle at the interface

- macroscopic picture: interplay of surface energies
- contributions from three possible interfaces: \( F = \gamma_{pl} S_{pl} + \gamma_{pg} S_{pg} + \gamma_{lg} S_{lg} \)
- rough estimate: undeformable flat interface \( \Rightarrow F(h) = \pi \gamma a^2 (h/a + \cos \theta_p)^2 \), where \( \cos \theta_p = (\gamma_{pg} - \gamma_{pl})/\gamma_{lg} \)

\[
\begin{array}{|c|c|}
\hline
a & \Delta F [k_B T] \\
\hline
10\text{nm} & 10^3 \\
100\text{nm} & 10^5 \\
1\mu m & 10^7 \\
\hline
\end{array}
\]
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\( h/a \)
Generic case: single particle at a flat interface

- particle pulled by the force \( f = \text{weight} - \text{buoyancy} \)
- interface effectively pinned by gravity at the distance \( \lambda = \sqrt{\gamma/\Delta \rho g} \) (capillary length)
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![Diagram showing a particle at a flat interface with angles $\theta_p$, $\beta$, and $\lambda$.]
Generic case: single particle at a flat interface

- the capillary equation for $|\nabla_\parallel u| \ll 1$ (balance of capillary and hydrostatic pressures across the interface)

$$-\gamma \nabla_\parallel^2 u + \frac{\gamma}{\lambda^2} u = 0$$

- the corresponding Green’s function $G(x, x') = G(|x, x'|)$ obeying the condition $G(r \to \infty) = 0$ reads

$$G(r) = \frac{1}{2\pi} K_0(r/\lambda) \sim \begin{cases} \ln(\lambda/r) & \text{for } r \ll \lambda \\ r^{-1/2} e^{-r/\lambda} & \text{for } r \gg \lambda \end{cases}$$
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Effective description, limit $\lambda \to \infty$

- particle replaced by an effective pressure distribution $\Pi(x)$
- for $\lambda \to \infty$ Poisson equation $-\gamma \nabla^2 u = \Pi(x)$
- in terms of complex variables $u(x) = \text{Re}V(z)$ with $V(z) = (2\pi\gamma)^{-1} \int d^2x' \Pi(z') \ln[\lambda/(z-z')]$
- for $\Pi(z')$ localized around the origin one can use the Taylor expansion

$$2\pi\gamma V(z) = \tilde{Q}_0 \ln(\lambda/z) + \sum_{n=1}^{\infty} \tilde{Q}_n n^{-1} z^{-n}$$

- with the multipoles $\tilde{Q}_n := \int d^2x' \Pi(z') z'^n = Q_n e^{i\phi_n}$ so that $Q_0 = \text{total external force}, Q_1 = \text{total external torque}; Q_{n \geq 2}$ correspond to free particles
- residue theorem $\Rightarrow$ all multipoles fully determined by the deformation around an arbitrary contour $C$ enclosing the origin: $\tilde{Q}_n = i\gamma \oint_C dz z^n (dV/dz)$
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Capillary interactions

- two particles at distance $d$, effective pressure $\Pi = \Pi_1 + \Pi_2$
- free energy
  \[ F = \int d^2 x \left[ \frac{\gamma}{2} (\nabla \parallel u)^2 - \Pi(x) u(x) \right] = -\frac{1}{2\gamma} \int d^2 x \int d^2 x' \Pi(x) G(x, x') \Pi(x') \]
- $F = F_{1,\text{self}} + F_{2,\text{self}} + \Delta F(d)$
- multipole expansion yields
  \[ \Delta F(d) = -\frac{1}{\gamma} \sum_{n=0}^{\infty} \sum_{n'=0}^{\infty} Q_{1,n} Q_{2,n'} g_{nn'} \cos(n\varphi_{1n} + n'\varphi_{2n'}) \times \begin{cases} \ln(\lambda/d) & n = n' = 0, \\ d^{n-n'} & \text{otherwise} \end{cases} \]
- in general $Q_{i,n} = Q_{i,n}(d)$ (feedback $u \rightarrow \Pi$), many-body interactions!
- but $Q_0$ and $Q_1$ can be fixed by external forces and torques
two particles at distance $d$, effective pressure $\Pi = \Pi_1 + \Pi_2$

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Spherical interfaces

- Assume small radial deformations $v(\Omega) = (r(\Omega) - R_0)/R_0$ and incompressibility of liquid $\Rightarrow$ free energy functional:

$$F[v(\Omega)] = \gamma R_0^2 \int_{\Omega_0} d\Omega \left[ \frac{1}{2} (\nabla_a v)^2 - v^2 - (\pi(\Omega) + \mu) v \right] + O(v^3, (\nabla_a v)^3)$$

- with $\int d\Omega v(\Omega) = 0$; condition $\delta F = 0$ yields $-\nabla^2_a v - 2v = \pi(\Omega) + \mu$

- Free energy $F = \min_{\{v(\Omega)\}} F$ in terms of the corresponding Green's function $G$ reads

$$F = -\frac{\gamma R_0^2}{2} \int d\Omega \int d\Omega' \; \pi(\Omega) G(\Omega, \Omega') \pi(\Omega')$$

- At small separations $G(\bar{\theta}) \xrightarrow{\bar{\theta} \to 0} - (2\pi)^{-1} \ln(\bar{\theta}) = -(2\pi)^{-1} \ln(r/R_0)$
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\mathcal{F}[\{\nu(\Omega)\}] = \gamma R_0^2 \int_{\Omega_0} d\Omega \left[ \frac{1}{2} (\nabla_a \nu)^2 - \nu^2 - (\pi(\Omega) + \mu) \nu \right] + O(\nu^3, (\nabla_a \nu)^3)
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Expansion in spherical harmonics

- **Capillary equation**
  \[ l(l+1) - 2 \nu_{lm} = \pi_{lm} + \mu \delta_{l0} \text{ with} \]
  \[ X_{lm} = \int d\Omega X(\Omega) Y_{lm}(\Omega) \]

- **\( l = 0 \): incompressibility** \( \nu_{00} = 0 \Rightarrow \mu = \pi_{00} \)

- **\( l = 1 \): translations** \( \nu_{1m} \) undefined, assume fixed center of mass \( \nu_{1m} = 0 \)

- **Free energy in terms of irreducible representation of rotation group**

  \[ \Delta F = -\gamma R_0^2 \sum_{l \geq 2} \sum_{m=-l}^{l} \sum_{m'=-l}^{l} \pi_{1,lm} \pi_{2,lm'} \frac{(-1)^{m'}}{l(l+1)-1} d_{ll}^{l,m} (\bar{\theta}) e^{i(m\phi_1 + m'\phi_2)} \]
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\]
in the limit $a, a' \ll R_0$ one has $\Delta F = \sum_{n, n'=0}^{\infty} \Delta F_{nn'}$ with $n = |m|$ and

\[
\Delta F_{nn'} = \gamma a^2 \frac{Q_{1,n} Q_{2,n'}}{(-2)^{n+n'+1} n! n'! \pi} \left( \frac{a}{R_0} \right)^{n+n'} \sum_{l \geq \max\{2, n, n'\}} \frac{(2l + 1)}{(l + 2)(l - 1)} \\
\times \left\{ \begin{array}{l}
\frac{(l + n')!}{(l - n)!} \left[ (-1)^n \cos(n\phi_1 + n'\phi_2) \cos \left( \frac{\bar{\theta}}{2} \right)^{n'-n} \sin \left( \frac{\bar{\theta}}{2} \right)^{n'+n} P_{l-n'}^{(n'+n, n'-n)}(\cos \bar{\theta}) \\
+ \cos(n\phi_1 - n'\phi_2) \left( \cos \left( \frac{\bar{\theta}}{2} \right)^{n'+n} \sin \left( \frac{\bar{\theta}}{2} \right)^{n'-n} P_{l-n'}^{(n'-n, n'+n)}(\cos \bar{\theta}) \right) \right], \quad n > 0, \quad n' > 0, \\
(-1)^n \cos(n\phi_1) P_l^n(\cos \bar{\theta}), \quad n > 0, \quad n' = 0, \\
2^{-1} P_l(\cos \bar{\theta}), \quad n = 0, \quad n' = 0,
\end{array} \right.
\]

$Q_{i,n}$ are capillary multipoles on the locally flat interface, i.e., defined on the plane tangent to the unit sphere at $\Omega_i$.

$R_0$ sets both the spatial separation and the capillary length.

more complex dependence on orientations
in the limit \(a, a' \ll R_0\) one has \(\Delta F = \sum_{n,n'=0}^{\infty} \Delta F_{nn'}\) with \(n = |m|\) and

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\]

\[
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(l + n')! \left[ (-1)^n \cos(n\phi_1 + n'\phi_2) \left(\cos\frac{\bar{\theta}}{2}\right)^{n'-n} \left(\sin\frac{\bar{\theta}}{2}\right)^{n'+n} P_{l-n'}^{(n'+n',n'-n)}(\cos\bar{\theta}) \\
+n \cos(n\phi_1 - n'\phi_2) \left(\cos\frac{\bar{\theta}}{2}\right)^{n'+n} \left(\sin\frac{\bar{\theta}}{2}\right)^{n'-n} P_{l-n'}^{(n'-n,n'+n)}(\cos\bar{\theta}) \\
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more complex dependence on orientations
Limit of small particles

- in the limit \( a, a' \ll R_0 \) one has \( \Delta F = \sum_{n,n'=0}^{\infty} \Delta F_{nn'} \) with \( n = |m| \) and

\[
\Delta F_{nn'} = \gamma a^2 \frac{Q_{1,n} Q_{2,n'}}{(-2)^{n+n'+1} n! n'! \pi} \left( \frac{a}{R_0} \right)^{n+n'} \sum_{l \geq \max\{2,n,n'\}} \frac{(2l+1)}{(l+2)(l-1)} \]

\[
\times \begin{cases} 
\frac{(l+n')!}{(l-n)!} \left[ (-1)^n \cos(n\phi_1 + n'\phi_2) \left( \cos \frac{\bar{\theta}}{2} \right)^{n'-n} \left( \sin \frac{\bar{\theta}}{2} \right)^{n'+n} P_{l-n'}^{(n'+n,n'-n)}(\cos \bar{\theta}) 
\right. \\
\left. + \cos(n\phi_1 - n'\phi_2) \left( \cos \frac{\bar{\theta}}{2} \right)^{n'+n} \left( \sin \frac{\bar{\theta}}{2} \right)^{n'-n} P_{l-n'}^{(n'-n,n'+n)}(\cos \bar{\theta}) \right] 
\end{cases} , \quad n > 0, \quad n' > 0,
\]

\[
(\frac{-1)^n}{2} \cos(n\phi_1) P_l^n(\cos \bar{\theta}), \quad 2^{-1} P_l(\cos \bar{\theta}),
\]

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(l - n)! \end{array} \right) \left[ \begin{array}{c} (-1)^n \cos(n \phi_1 + n' \phi_2) \left( \cos \frac{\bar{\theta}}{2} \right)^{n'+n} \left( \sin \frac{\bar{\theta}}{2} \right)^{n'+n} P_{l-n'}^{(n'+n,n'-n)}(\cos \bar{\theta}) \\
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(-1)^n \cos(n\phi_1) P^n_l(\cos \bar{\theta}), \quad n > 0, \quad n' = 0, \\
2^{-1} P^n_l(\cos \bar{\theta}), \quad n = 0, \quad n' = 0,
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more complex dependence on orientations
Numerical calculations

- surface free energy minimized by using software Surface Evolver based on the gradient descent method

- minimized expression:

\[
\mathcal{F}[\{r(\Omega)\}, h_i, \psi_i; \bar{\theta}, \phi_i, f_i, T_i, \theta_{p,i}, a_i, V_i, \lambda_0] = \\
= \gamma S_{lg} + \sum_{i=1,2} (-\gamma \cos \theta_{p,i} S_{pl,i} - f_i h_i - T_i \cdot \psi_i) - \lambda_0 (V - V_i).
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\]
Results: monopoles

- smooth spherical particles, external radial forces $f = \gamma a Q_0$, fixed CM

\[
\frac{\Delta F_{00}(\bar{\theta})}{\gamma a^2} = -Q_0^2 G(\bar{\theta}) = \frac{Q_0^2}{4\pi} \left[ \frac{1}{2} + \frac{4}{3} \cos \bar{\theta} + 2 \cos \bar{\theta} \ln \left( \sin \frac{\bar{\theta}}{2} \right) \right]
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\]

\[
\Delta F_{00}/(\gamma a^2 Q_0^2)
\]

- $R_0/a = 4$
- $R_0/a = 6$
- $R_0/a = 8$
Results: dipoles

- three metastable branches for three different orientational configurations

\[
\Delta F_{11}(\bar{\theta}, \phi_1, \phi_2) = \gamma a^2 \frac{Q_1^2}{8\pi} \left( \frac{a}{R_0} \right)^2 \begin{cases} 
- f_+ (\bar{\theta}) + f_- (\bar{\theta}), & \text{for } \bar{\theta} < \bar{\theta}_0, \quad \uparrow \uparrow \\
- f_+ (\bar{\theta}) - f_- (\bar{\theta}), & \text{for } \bar{\theta}_0 < \bar{\theta} < \bar{\theta}_1, \quad \leftrightarrow \\
f_+ (\bar{\theta}) - f_- (\bar{\theta}), & \text{for } \bar{\theta} > \bar{\theta}_1, \quad \uparrow \downarrow 
\end{cases}
\]

- where \( f_- (\bar{\theta}_0) = 0 \) and \( f_+ (\bar{\theta}_1) = 0 \) and

\[
f_+ (\theta) := \frac{1}{\sin^2(\theta/2)} - 4 \sin^2 \frac{\theta}{2} \ln \left( \sin \frac{\theta}{2} \right) - \frac{20}{3} \sin^2 \frac{\theta}{2} + 2,
\]

\[
f_- (\theta) := 4 \left( \cos \frac{\theta}{2} \right)^2 \ln \left( \sin \frac{\theta}{2} \right) + \frac{20}{3} \cos^2 \frac{\theta}{2}
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\]
Results: dipoles

• pinned contact lines, external torques \( T = \gamma a^2 Q_1 \), fixed CM
Results: free spheroidal particles

- free, smooth prolate spheroids; approximation:
  \[ Q_2 = Q_2(R_0) \simeq 2\pi \Delta r|_{\theta=a/R_0/a} \]

\[ \Delta F_{22}(\bar{\theta}, \phi_1, \phi_2) = -\gamma a^2 \frac{3Q_2^2}{64\pi} \left( \frac{a}{R_0} \right)^4 \frac{1}{\sin^4(\bar{\theta}/2)}. \]
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\]
free energy depends on the contact angle $\theta_0$ and boundary conditions of either a free ($\sigma = A$) or a pinned ($\sigma = B$) contact line at the substrate.
Sessile drops: free energy

- after subtracting self-energies $F_{i,\text{self}}$ one gets

$$\Delta F^{(N)}_\sigma := F^{(N)}_\sigma(\Omega_1, \ldots, \Omega_N, \theta_0) - \sum_{i=1}^{N} F_{i,\text{self}} =$$

$$= \sum_{i=1}^{N} \Delta F^{(1)}_\sigma(\theta_i, \theta_0) + \sum_{i<j} V_\sigma(\Omega_i, \Omega_j, \theta_0)$$

- substrate potential $\Delta F^{(1)}_\sigma$ and pair-potential $V_\sigma$:

$$\Delta F^{(1)}_\sigma = -\frac{f_i^2}{2\gamma} [G_{\sigma,\text{reg}}(\Omega_i, \Omega_i) - G_{\sigma,\text{reg}}(0, 0)]$$

$$V_\sigma = -\frac{f_i f_j}{2\gamma} [G_\sigma(\Omega_i, \Omega_j) + G_\sigma(\Omega_j, \Omega_i)]$$
Sessile drops: free energy

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$$
\Delta F^{(N)} \sigma := \left[ F^{(N)}_\sigma (\Omega_1, \ldots, \Omega_N, \theta_0) - \sum_{i=1}^{N} F_{i,\text{self}} \right] = \\
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$$

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$$
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$$
for $\Omega \in \Omega_0$ Green’s functions $G_\sigma$ satisfy

$$-(\nabla^2 + 2)G_\sigma(\Omega, \Omega', \theta_0) = \delta(\Omega, \Omega') + \Delta_\sigma(\Omega, \Omega', \theta_0)$$

functions $\Delta_\sigma(\Omega, \Omega', \theta_0)$ corresponding to $\mu$ and $\pi_{CM}$ determined from the force balance and incompressibility condition $\int_{\Omega_0} d\Omega \ G_\sigma(\Omega, \Omega') = 0$

with boundary conditions:

$$((\sin \theta_0 \partial_\theta G_A(\Omega, \Omega') - \cos \theta_0 G_A(\Omega, \Omega'))|_{\Omega \in \partial \Omega_0} = 0, \ G_B(\Omega, \Omega')|_{\Omega \in \partial \Omega_0} = 0.$$
for \( \Omega \in \Omega_0 \) Green’s functions \( G_\sigma \) satisfy

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\]
Sessile drops: Green’s functions

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  G_B(\Omega, \Omega')|_{\Omega \in \partial \Omega_0} = 0.
  \]
Sessile drops: special case $\theta_0 = \pi/2$

- $f$ images

**Free c.l.**

**Pinned c.l.**
Sessile drops: special case $\theta_0 = \pi/2$

$$\gamma \Delta F^{(2)}_A / (f_1 f_2) =$$

(a) \hspace{1cm} (b) \hspace{1cm} (c) \hspace{1cm} (d)

(e) \hspace{1cm} (f) \hspace{1cm} (g) \hspace{1cm} (h)
Sessile drops: special case $\theta_0 = \pi/2$

$$\gamma \Delta F_B^{(2)}/(f_1 f_2) =$$
interactions between **monopoles** and **dipoles** on spherical interface are non-monotonic and much different than on a flat interface

interactions between **spheroids** are quite similar

importance of **curvature** only in case of external fields

the effects of **boundary conditions** on the substrate for monopoles are long-ranged and independent of $R_0$

the effective **confining potential** depends qualitatively on the boundary conditions

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