

Spontaneous motion and deformation of a self-propelled droplet: Supplementary information

Natsuhiko Yoshinaga^{1,*}

¹ WPI - Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

S1. PERTURBED CONCENTRATION

Here we show the explicit forms appearing in the expansion (41) of a concentration field close to the critical point of drift bifurcation. The second-order in the expansion is expressed in terms of a normal vector and a surface gradient derivative as

$$\begin{aligned}
& c_I^{(2)}(\mathbf{r}_G + \mathbf{s}) \\
&= \frac{A}{D^3} \int_{\mathbf{q}} (i\mathbf{q} \cdot \hat{\mathbf{u}}) G_q^3 S_q e^{-i\mathbf{q} \cdot \mathbf{s}} + \frac{A}{D^3} \int_{\mathbf{q}} (i\mathbf{q} \cdot \mathbf{u})^2 G_q^3 S_q e^{-i\mathbf{q} \cdot \mathbf{s}} \\
&= -\dot{u}_i \frac{A}{D^3} \left[n_i \left(\frac{\partial Q_3^{(0)}(s)}{\partial s} + \frac{\partial Q_3^{(1)}(\theta, \varphi)}{\partial s} \right) + \nabla_{s,i} Q_3^{(1)}(\theta, \varphi) \right] \\
&\quad + u_i u_j \frac{A}{D^3} \left[\frac{1}{R_0} \mathcal{P}_{ij} \frac{\partial}{\partial s} + n_i n_j \frac{\partial^2}{\partial s^2} \right] \left(Q_3^{(0)}(s) + Q_3^{(1)} \right) + u_i u_j \frac{A}{D^3} \left[(n_j \nabla_{s,i} + n_i \nabla_{s,j}) \frac{\partial Q_3^{(1)}}{\partial s} + \nabla_{s,i} \nabla_{s,j} Q_3^{(1)} \right]. \quad (\text{S1.1})
\end{aligned}$$

Similarly, the third-order in the expansion is expressed as

$$\begin{aligned}
& c_I^{(3)}(\mathbf{r}_G + \mathbf{s}) \\
&= -\frac{A}{D^4} \int_{\mathbf{q}} (i\mathbf{q} \cdot \mathbf{u})^3 G_q^4 S_q e^{-i\mathbf{q} \cdot \mathbf{s}} \\
&= u_i u_j u_k \frac{A}{D^4} \left[\left(-\frac{1}{R_0^2} (\llbracket \delta_{ij} n_k \rrbracket - 3n_i n_j n_k) \frac{\partial}{\partial s} + \frac{1}{R_0} (\llbracket \delta_{jk} n_i \rrbracket - 3n_i n_j n_k) \frac{\partial^2}{\partial s^2} + n_i n_j n_k \frac{\partial^3}{\partial s^3} \right) \left(Q_4^{(0)}(s) + Q_4^{(1)}(\theta, \varphi) \right) \right. \\
&\quad \left. + \frac{1}{R_0} \llbracket \mathcal{P}_{ij} \nabla_{s,k} \rrbracket \frac{\partial Q_4^{(1)}}{\partial s} + \llbracket n_i n_j \nabla_{s,k} \rrbracket \frac{\partial^2 Q_4^{(1)}}{\partial s^2} + \llbracket n_j \nabla_{s,i} \nabla_{s,k} \rrbracket \frac{\partial Q_4^{(1)}}{\partial s} + \llbracket \nabla_{s,i} \nabla_{s,j} \nabla_{s,k} \rrbracket Q_4^{(1)} \right], \quad (\text{S1.2})
\end{aligned}$$

where $\llbracket \quad \rrbracket$ shows two other permutations among i, j, k .

S2. COEFFICIENTS OF THE AMPLITUDE EQUATIONS

A. first mode

Using the concentration field obtained in the main text (41), the velocity resulting from the normal force becomes

$$u_{i,1}^{(\alpha)} = \sum_{\alpha=0}^5 \sum_{\beta=0}^3 u_{i,1}^{(\alpha,\beta)} \quad (\text{S2.1})$$

where $\alpha = 0$ is the isotropic part in (28) and (31). For any α , the zeroth-order contribution in the expansion of the concentration field vanishes. This is because there is no $l = 1$ mode in deformation.

$$u_{i,1}^{(\alpha,0)} = u_{i,2}^{(\alpha,0)} = 0. \quad (\text{S2.2})$$

*E-mail: yoshinaga@wpi-aimr.tohoku.ac.jp

For $\alpha \geq 1$

$$u_{i,1}^{(\alpha,2)}, u_{i,2}^{(\alpha,2)} = \mathcal{O}(\dot{u}_j S_{ij}, u_j u_k T_{ijk}), \quad (\text{S2.3})$$

$$u_{i,1}^{(\sigma,3)}, u_{i,2}^{(\sigma,3)} = \mathcal{O}(u_i u_j u_k S_{jk}, u_j u_k u_l D_{ijkl}), \quad (\text{S2.4})$$

and we may neglect them. The results are summarized.

$$u_{i,1}^{(0,1)} = -\frac{8\gamma_c A}{15D^2\eta} \left[u_i \frac{\partial Q_2^{(0)}}{\partial s} + u_j S_{ij} \left(a_{2,0}^{(2)} \frac{\partial \bar{Q}_2^{(1)}}{\partial s} + a_{1,1}^{(2)} \bar{Q}_2^{(1)} \right) \right]. \quad (\text{S2.5})$$

using (D2) and (D6).

$$u_{i,1}^{(0,2)} = \frac{8\gamma_c A}{15D^3\eta} \left[\dot{u}_i \frac{\partial Q_3^{(0)}}{\partial s} + \dot{u}_j S_{ij} \left(a_{2,0}^{(2)} \frac{\partial \bar{Q}_{3,2}^{(1)}}{\partial s} + a_{1,1}^{(2)} \bar{Q}_{3,2}^{(1)} \right) \right], \quad (\text{S2.6})$$

$$u_{i,1}^{(0,3)} = \frac{8\gamma_c A}{15D^4\eta} u_i u^2 \left[-\frac{6}{5R_0^2} \frac{\partial}{\partial s} + \frac{6}{5R_0} \frac{\partial^2}{\partial s^2} + \frac{3}{5} \frac{\partial^3}{\partial s^3} \right] Q_4^{(0)}(s). \quad (\text{S2.7})$$

Here we have used (D2) and (D3). At linear order in deformation, the other terms have only isotropic part of Q_n . The results of $u_{i,1}^{(\alpha,1)}$ are listed:

$$u_{i,1}^{(1,1)} = -\frac{8\gamma_c A}{5R_0\eta D^2} u_j a_{2,0}^{(2)} \frac{\partial Q_2^{(0)}(s)}{\partial s} S_{ij}. \quad (\text{S2.8})$$

$$u_{i,1}^{(2,1)} \simeq \frac{2\gamma_c A}{5\eta D^2} a_{1,1}^{(2)} u_j S_{ij} \frac{\partial Q_2^{(0)}}{\partial s}, \quad (\text{S2.9})$$

$$u_{\alpha,1}^{(3,1)} = \frac{6\gamma_c A}{15\eta D^2} a_{1,1}^{(2)} u_j S_{ij} \frac{\partial Q_2^{(0)}}{\partial s}, \quad (\text{S2.10})$$

$$u_{i,1}^{(4,1)} = -\frac{16\gamma_c A}{15\eta R_0 D^2} a_{2,0}^{(2)} u_j S_{ij} \frac{\partial Q_2^{(0)}}{\partial s}, \quad (\text{S2.11})$$

$$u_{i,1}^{(5,1)} = -\frac{16A\gamma_c}{15D^2 R_0 \eta} a_{2,0}^{(2)} \frac{\partial Q_2^{(0)}}{\partial s} u_j S_{ij}. \quad (\text{S2.12})$$

The velocity originated from tangential force is decomposed with

$$\begin{pmatrix} u_{i,2}^{(0)} \\ u_{i,2}^{(1)} \\ u_{i,2}^{(2)} \\ u_{i,2}^{(3)} \\ u_{i,2}^{(4)} \\ u_{i,2}^{(4)} \end{pmatrix} = \frac{\gamma_c R_0}{\Omega} \int da \int da' n_i(a) \mathcal{T}_{jk}(\mathbf{r}, \mathbf{r}(a')) \left\{ \begin{array}{l} \mathcal{P}_{k\alpha}(a') \nabla_\alpha n_j(a) \\ \frac{3\gamma_c}{\Omega} \mathcal{P}_{k\alpha}(a') \nabla_\alpha \delta R(a) n_j(a) \\ -\frac{\gamma_c R_0}{\Omega} \nabla_{s,j} \delta R(a) \mathcal{P}_{k\alpha}(a') \nabla_\alpha \\ \frac{\gamma_c R_0}{\Omega} n_j(a) \left[n_k^{(0)}(a') \nabla_{s,\alpha} \delta R(a') + n_\alpha^{(0)}(a') \nabla_{s,k} \delta R(a') \right] \nabla_\alpha \\ \frac{2\gamma_c}{\Omega} n_j(a) \mathcal{P}_{k\alpha}(a') \nabla_\alpha c(a', t) \delta R(a') \end{array} \right\} c(a', t) \quad (\text{S2.13})$$

As the contribution from normal force (30), $u_{i,2}^{(0)}$, $u_{i,2}^{(3)}$, and $u_{i,2}^{(4)}$ are readily calculated as

$$\begin{pmatrix} u_{i,2}^{(0)} \\ u_{i,2}^{(3)} \\ u_{i,2}^{(4)} \end{pmatrix} = \frac{\gamma_c R_0^2}{5\Omega\eta} \int da' \left\{ \begin{array}{l} \mathcal{P}_{i\alpha}(a') \nabla_\alpha \\ \left[\frac{4}{3} n_i^{(0)}(a') \nabla_{s,\alpha} \delta R(a') + n_\alpha^{(0)}(a') \nabla_{s,i} \delta R(a') \right] \nabla_\alpha \\ \frac{2}{R_0} \mathcal{P}_{i\alpha}(a') \nabla_\alpha c(a', t) \delta R(a') \end{array} \right\} c(a', t) \quad (\text{S2.14})$$

Here the projection onto the normal vector of a spherical surface is

$$\mathcal{P}_{jk}(a') = \delta_{jk} - n_j(a') n_k(a'), \quad (\text{S2.15})$$

which satisfies $n_j(a') \mathcal{P}_{jk}(a') = n_k(a') \mathcal{P}_{jk}(a') = 0$.

Each term is further expanded following (41) as (S2.1):

$$u_{i,2}^{(\alpha)} = \sum_{\alpha=0}^4 \sum_{\beta=0}^3 u_{i,2}^{(\alpha,\beta)}. \quad (\text{S2.16})$$

Similar to $u_{i,1}$, we obtain

$$u_{i,2}^{(0,1)} = \frac{2\gamma_c A}{5D^2\eta} u_i \frac{\partial Q_2^{(0)}}{\partial s} + \frac{\gamma_c A}{5D^2\eta} u_j S_{ij} \left[-a_{2,0}^{(2)} \frac{\partial \bar{Q}_{2,2}^{(1)}}{\partial s} + R_0 a_{1,1}^{(2)} \frac{\partial \bar{Q}_{2,2}^{(1)}}{\partial s} + R_0 a_{0,2}^{(2)} \bar{Q}_{2,2}^{(1)} \right], \quad (\text{S2.17})$$

$$u_{i,2}^{(0,2)} = -\frac{2\gamma_c A}{5D^3\eta} \dot{u}_i \frac{\partial Q_3^{(0)}}{\partial s} - \frac{\gamma_c A}{5D^2\eta} \dot{u}_j S_{ij} \left[-a_{2,0}^{(2)} \frac{\partial \bar{Q}_{2,2}^{(1)}}{\partial s} + R_0 a_{1,1}^{(2)} \frac{\partial \bar{Q}_{2,2}^{(1)}}{\partial s} + R_0 a_{0,2}^{(2)} \bar{Q}_{2,2}^{(1)} \right], \quad (\text{S2.18})$$

$$u_{i,2}^{(0,3)} = \frac{2\gamma_c A}{5D^4\eta} u^2 u_i \left[-\frac{6}{5R_0^2} \frac{\partial Q_4^{(0)}}{\partial s} + \frac{6}{5R_0} \frac{\partial^2 Q_4^{(0)}}{\partial s^2} + \frac{3}{5} \frac{\partial^3 Q_4^{(0)}}{\partial s^3} \right] \quad (\text{S2.19})$$

$$u_{i,2}^{(1,1)} = \frac{6A\gamma_c}{5D^2 R_0 \eta} a_{2,0}^{(2)} \frac{\partial Q_2^{(0)}}{\partial s} u_j S_{ij} \quad (\text{S2.20})$$

$$u_{i,2}^{(2,1)} = -\frac{7\gamma_c A}{15\eta D} u_j S_{ij} a_{1,1}^{(2)} \frac{\partial Q_2^{(0)}}{\partial s}, \quad (\text{S2.21})$$

$$u_{i,2}^{(3,1)} = \frac{\gamma_c R_0 A}{15\eta D^2} a_{1,1}^{(2)} u_j S_{ij} \left(\frac{4}{R_0} \frac{\partial Q_2^{(0)}}{\partial s} + 3 \frac{\partial^2 Q_2^{(0)}}{\partial s^2} \right), \quad (\text{S2.22})$$

$$u_{i,2}^{(4,1)} = -\frac{2\gamma_c A}{5\eta D^2} a_{2,0}^{(2)} u_j S_{ij} \frac{\partial Q_2^{(0)}}{\partial s}. \quad (\text{S2.23})$$

B. second mode

We expand the right-hand side of (51) with the following terms; the contribution from normal force is

$$\begin{Bmatrix} M_{ij}^{(1,1)} \\ M_{ij}^{(1,2)} \\ M_{ij}^{(1,3)} \\ M_{ij}^{(1,4)} \end{Bmatrix} = \frac{\gamma_c}{\Omega} \int da \int da' \overline{n_i(a) n_j(a)} \overline{n_j(a') n_i(a')} \mathbb{T}_{\alpha\beta}(\mathbf{r}(a), \mathbf{r}(a')) \begin{Bmatrix} R_0 n_\alpha(a) n_\beta(a') \kappa(a') \\ 2\nabla_{s,\alpha} \delta R(a) n_\beta(a') \\ -R_0 \nabla_{s,\beta} \delta R(a') n_\alpha(a) \kappa(a') \\ 4\delta R(a') n_\alpha^{(0)}(a) n_\beta^{(0)}(a') \end{Bmatrix} c(a') \quad (\text{S2.24})$$

and the contribution from tangential force is

$$\begin{Bmatrix} M_{ij}^{(2,1)} \\ M_{ij}^{(2,2)} \\ M_{ij}^{(2,3)} \\ M_{ij}^{(2,4)} \end{Bmatrix} = \frac{\gamma_c R_0}{\Omega} \int da \int da' \overline{n_i(a) n_j(a)} \overline{n_j(a') n_i(a')} \mathbb{T}_{\alpha\beta}(\mathbf{r}(a), \mathbf{r}(a')) \begin{Bmatrix} n_\alpha(a) \mathcal{P}_{\beta\gamma}(a') \\ -\nabla_{s,\alpha} \delta R(a) \mathcal{P}_{\beta\gamma}(a') \\ n_\alpha(a) [\nabla_{s,\beta} \delta R(a') n_\gamma(a') + \nabla_{s,\gamma} \delta R(a') n_\beta(a')] \\ 2n_\alpha(a) \mathcal{P}_{\beta\gamma}(a') \frac{\delta R(a')}{R_0} \end{Bmatrix} (\nabla_\gamma c(a'))_I.$$

These are simplified as

$$\begin{Bmatrix} M_{ij}^{(1,1)} \\ M_{ij}^{(1,3)} \\ M_{ij}^{(1,4)} \end{Bmatrix} = \frac{4\gamma_c R_0^2}{35\Omega\eta} \int da' \begin{Bmatrix} \overline{n_i(a) n_j(a)} \kappa(a') \\ \frac{35\eta}{2R_0^2} \mathcal{B}_2^{(3)} (\nabla_{s,i} \delta R(a') n_j(a') + \nabla_{s,j} \delta R(a') n_i(a')) \\ -\frac{4}{R_0^2} \overline{n_i(a') n_j(a')} \delta R(a') \end{Bmatrix} c(a'). \quad (\text{S2.25})$$

$$\begin{Bmatrix} M_{ij}^{(2,1)} \\ M_{ij}^{(2,3)} \\ M_{ij}^{(2,4)} \end{Bmatrix} = \frac{\gamma_c R_0}{\Omega} \int da' \begin{Bmatrix} \mathcal{B}_2^{(3)} (\delta_{i\gamma} n_j(a') + \delta_{j\gamma} n_i(a') - 2n_i(a') n_j(a') n_\gamma(a')) \\ \left[\mathcal{B}_2^{(3)} (n_j(a') \nabla_{s,i} \delta R(a') + n_i(a') \nabla_{s,j} \delta R(a')) n_\gamma(a') + \frac{4R_0}{35\eta} \overline{n_i(a') n_j(a')} \nabla_{s,\gamma} \delta R(a') \right] \\ \frac{2}{R_0} \mathcal{B}_2^{(3)} (\delta_{i\gamma} n_j(a') + \delta_{j\gamma} n_i(a') - 2n_i(a') n_j(a') n_\gamma(a')) \delta R(a') \end{Bmatrix} (\nabla_\gamma c(a'))_I$$

These are further expanded according to (41) for $\alpha = 1, 2, 3, 4$ and $k = 1, 2$ as

$$M_{ij}^{(k,\alpha)} = M_{ij}^{(k,\alpha,0)} + M_{ij}^{(k,\alpha,1)} + M_{ij}^{(k,\alpha,2)} + M_{ij}^{(k,\alpha,3)} + \dots \quad (\text{S2.26})$$

For all α and $k = 1, 2$

$$M_{ij}^{(k,\alpha,1)} = \mathcal{O}(u_k T_{ijk}), \quad (\text{S2.27})$$

$$M_{ij}^{(k,\alpha,3)} = \mathcal{O}(u_i u_k u_l T_{ijkl}) \quad (\text{S2.28})$$

and for $\alpha \geq 2$, we obtain

$$M_{ij}^{(k,\alpha,2)} = \mathcal{O}(u_k u_l D_{ijkl}). \quad (\text{S2.29})$$

The expressions of these terms are listed here.

$$M_{ij}^{(1,1,0)} = -\frac{4A\gamma_c R_0}{35D\eta} a_{2,0}^{(2)} \left(\frac{4}{R_0^2} Q_1^{(0)}(s) + \frac{2}{R_0} \bar{Q}_{1,2}^{(1)}(s) \right) S_{ij}. \quad (\text{S2.30})$$

$$M_{ij}^{(1,1,2)} = -\frac{16\gamma_c A}{175D^3\eta} \frac{u_i u_j}{u_i u_j} \left(-\frac{1}{R_0} \frac{\partial Q_3^{(0)}}{\partial s} + \frac{\partial^2 Q_3^{(0)}}{\partial s^2} \right), \quad (\text{S2.31})$$

$$M_{ij}^{(1,2,0)} = 0, \quad (\text{S2.32})$$

$$M_{ij}^{(1,3,0)} = \frac{4A\gamma_c}{DR_0} \mathcal{B}_2^{(3)} a_{1,1}^{(2)} S_{ij} Q_1^{(0)}, \quad (\text{S2.33})$$

$$M_{ij}^{(1,4,0)} = -\frac{16\gamma_c A}{35R_0\eta D} a_{2,0}^{(2)} Q_1^{(0)} S_{ij}, \quad (\text{S2.34})$$

$$M_{ij}^{(2,1,0)} = \frac{2\gamma_c A}{D} \mathcal{B}_2 a_{1,1}^{(2)} \bar{Q}_{1,2} S_{ij}, \quad (\text{S2.35})$$

$$M_{ij}^{(2,1,2)} = \frac{12\gamma_c A}{5R_0 D^3} \mathcal{B}_2^{(3)} \frac{u_i u_j}{u_i u_j} \left[-\frac{1}{R_0} \frac{\partial Q_3^{(0)}}{\partial s} + \frac{\partial^2 Q_3^{(0)}}{\partial s^2} \right], \quad (\text{S2.36})$$

$$M_{ij}^{(2,2,0)} = 0, \quad (\text{S2.37})$$

$$M_{ij}^{(2,3,0)} = \frac{2\gamma_c A}{D} \mathcal{B}_2^{(3)} a_{1,1}^{(2)} \frac{\partial Q_1^{(0)}}{\partial s} S_{ij}, \quad (\text{S2.38})$$

$$M_{ij}^{(2,4,0)} = 0. \quad (\text{S2.39})$$

C. third mode

Similar to the second mode, we expand the right-hand side of (56) with the following terms:

$$\left\{ \begin{array}{l} N_{ijk}^{(1,1)} \\ N_{ijk}^{(1,2)} \\ N_{ijk}^{(1,3)} \\ N_{ijk}^{(1,4)} \end{array} \right\} = \frac{\gamma_c R_0}{\Omega} \int da \int da' \overline{n_i(a) n_j(a) n_k(a)} \mathbb{T}_{\alpha\beta}(\mathbf{r}(a), \mathbf{r}(a')) \left\{ \begin{array}{l} n_\alpha(a) n_\beta(a') \kappa(a') \\ \frac{2}{R_0} \nabla_{s,\alpha} \delta R(a) n_\beta(a') \\ -\nabla_{s,\beta} \delta R(a') n_\alpha(a) \kappa(a') \\ -\frac{4}{R_0} n_\alpha^{(0)}(a) n_\beta^{(0)}(a') \frac{\delta R(a')}{R_0} \end{array} \right\} c(a') \quad (\text{S2.40})$$

for the contribution from normal force, and

$$\left\{ N_{ijk}^{(2,\alpha)} \right\} = \frac{\gamma_c R_0}{\Omega} \int da \int da' \overline{n_i(a) n_j(a) n_k(a)} \mathbb{T}_{\alpha\beta}(\mathbf{r}(a), \mathbf{r}(a')) \left\{ \begin{array}{l} n_\alpha(a) \mathcal{P}_{\beta\gamma}(a') (\nabla_\gamma c(a'))_I \\ -\nabla_{s,\alpha} \delta R(a) \mathcal{P}_{\beta\gamma}(a') (\nabla_\gamma c(a'))_I \\ n_\alpha(a) [\nabla_{s,\beta} \delta R(a') n_\gamma(a') + \nabla_{s,\gamma} \delta R(a') n_\beta(a')] (\nabla_\gamma c(a'))_I \\ 2n_\alpha^{(0)}(a) \mathcal{P}_{\beta\gamma}(a') \frac{\delta R(a')}{R_0} (\nabla_\gamma c(a'))_I \end{array} \right\} \quad (\text{S2.41})$$

for the contribution from tangential force. These are again simplified as

$$\begin{Bmatrix} N_{ijk}^{(1,1)} \\ N_{ijk}^{(1,3)} \\ N_{ijk}^{(1,4)} \end{Bmatrix} \simeq \frac{8\gamma_c R_0^2}{105\eta\Omega} \int da' \left\{ \begin{array}{l} \overline{n_i(a')n_j(a')n_k(a')\kappa(a')} \\ \frac{1}{4R_0}\overline{\delta_{i\beta}n_j(a')n_k(a')\nabla_{s,\beta}\delta R(a')} \\ -\frac{4}{R_0^2}\overline{n_i(a')n_j(a')n_k(a')\delta R(a')} \end{array} \right\} c(a') \quad (\text{S2.42})$$

and

$$\begin{Bmatrix} N_{ijk}^{(2,1)} \\ N_{ijk}^{(2,3)} \\ N_{ijk}^{(2,4)} \end{Bmatrix} \simeq \frac{\gamma_c R_0^2}{105\eta\Omega} \int da' \left\{ \begin{array}{l} \overline{\delta_{i\beta}n_j(a')n_k(a')\mathcal{P}_{\beta\gamma}(a')\nabla_\gamma c(a')} \\ \overline{\delta_{i\beta}n_j(a')n_k(a')\nabla_{s,\beta}\delta R(a')n_\gamma(a')\nabla_\gamma c(a')} + 8 \int da' \overline{n_i(a')n_j(a')n_k(a')\nabla_{s,\gamma}\delta R(a')\nabla_\gamma c(a')} \\ \frac{2}{R_0}\overline{\delta_{i\beta}n_j(a')n_k(a')\mathcal{P}_{\beta\gamma}(a')\delta R(a')\nabla_\gamma c(a')} \end{array} \right\} \quad (\text{S2.43})$$

where

$$\overline{\delta_{i\beta}n_j(a')n_k(a')} = \delta_{i\beta}n_j(a')n_k(a') + \delta_{j\beta}n_i(a')n_k(a') + \delta_{k\beta}n_i(a')n_j(a') - \frac{1}{5}(\delta_{ij}\delta_{k\beta} + \delta_{ik}\delta_{j\beta} + \delta_{jk}\delta_{i\beta}). \quad (\text{S2.44})$$

These are further expanded similar to (S2.26). The results are summarized here.

$$N_{ijk}^{(1,1,0)} \simeq -\frac{8A\gamma_c}{105\eta D} a_{3,0}^{(3)} \left[2\overline{Q_{1,3}^{(1)}} + \frac{10}{R_0}Q_1^{(0)} \right] T_{ijk}, \quad (\text{S2.45})$$

$$N_{ijk}^{(1,1,1)} = -\frac{16\gamma_c A}{735\eta D^2} \left(\frac{\partial \overline{Q_{2,2}^{(1)}}}{\partial s} + \frac{2}{R_0} \frac{\partial Q_2^{(0)}}{\partial s} - \frac{2}{R_0} \overline{Q_{2,2}^{(1)}} \right) \overline{S_{ij}u_k}, \quad (\text{S2.46})$$

$$N_{ijk}^{(1,1,3)} \simeq -\frac{32\gamma_c A}{35^2\eta D^4} \overline{u_i u_j u_k} \left(\frac{3}{R_0^2} \frac{\partial Q_4^{(0)}}{\partial s} - \frac{3}{R_0} \frac{\partial^2 Q_4^{(0)}}{\partial s^2} + \frac{\partial^3 Q_4^{(0)}}{\partial s^3} \right), \quad (\text{S2.47})$$

$$N_{ijk}^{(1,2,0)} = 0, \quad (\text{S2.48})$$

$$N_{ijk}^{(1,2,1)} = -\frac{4\gamma_c A}{35D^2\eta} \frac{1}{R_0} \frac{\partial Q_2^{(0)}}{\partial s} a_{2,0}^{(2)} \overline{u_k S_{ij}}, \quad (\text{S2.49})$$

$$N_{ijk}^{(1,3,0)} \simeq \frac{2\gamma_c A}{35\eta D} a_{2,1}^{(3)} Q_1^{(0)} T_{ijk}, \quad (\text{S2.50})$$

$$N_{ijk}^{(1,3,1)} = \frac{16\gamma_c A}{735D^2\eta} a_{2,0}^{(2)} \frac{1}{R_0} \frac{\partial Q_2^{(0)}}{\partial s} \overline{u_k S_{ij}}, \quad (\text{S2.51})$$

$$N_{ijk}^{(1,4,0)} = -\frac{32\gamma_c A}{105\eta D R_0} a_{3,0}^{(3)} T_{ijk} Q_1^{(0)}, \quad (\text{S2.52})$$

$$N_{ijk}^{(1,4,1)} = -\frac{32\gamma_c A}{105\eta D^2 R_0} u_\alpha \left[a_{4,0}^{(4)} D_{ijk\gamma} + \frac{1}{7} a_{2,0}^{(2)} \overline{S_{ij} \delta_{k\gamma}} \right] \frac{\partial Q_2^{(0)}}{\partial s}, \quad (\text{S2.53})$$

$$N_{ijk}^{(2,1,0)} = \frac{\gamma_c A R_0}{35\eta D} a_{2,1}^{(3)} T_{ijk} \overline{Q_{1,3}^{(1)}}, \quad (\text{S2.54})$$

$$N_{ijk}^{(2,1,1)} = \frac{4\gamma_c A}{735\eta D^2} a_{2,0}^{(2)} \left(3 \frac{\partial \overline{Q_{2,3}^{(1)}}}{\partial s} - \frac{6}{R_0} \overline{Q_{2,3}^{(1)}} \right) \overline{u_k S_{ij}} \quad (\text{S2.55})$$

$$N_{ijk}^{(2,1,3)} = \frac{24\gamma_c A}{35^2\eta D^4} \overline{u_i u_j u_k} \left(\frac{3}{R_0^2} \frac{\partial Q_4^{(0)}}{\partial s} - \frac{3}{R_0} \frac{\partial^2 Q_4^{(0)}}{\partial s^2} + \frac{\partial^3 Q_4^{(0)}}{\partial s^3} \right) \quad (\text{S2.56})$$

$$N_{ijk}^{(2,2,0)} = 0, \quad (\text{S2.57})$$

$$N_{ijk}^{(2,2,1)} = \frac{2\gamma_c A}{15\eta D^2} a_{2,0}^{(2)} \overline{S_{ij} u_k} \frac{1}{R_0} \frac{\partial Q_2^{(0)}}{\partial s}, \quad (\text{S2.58})$$

$$N_{ijk}^{(2,3,0)} = \frac{\gamma_c R_0 A}{35\eta D} a_{2,1}^{(3)} \frac{\partial Q_1^{(0)}}{\partial s} T_{ijk}, \quad (\text{S2.59})$$

$$N_{ijk}^{(2,3,1)} = \frac{8\gamma_c A}{735 D^2} a_{2,0}^{(2)} \overline{S_{jk} u_i} \left[\frac{\partial^2 Q_2^{(0)}}{\partial s^2} - \frac{2}{R_0} \frac{\partial Q_2^{(0)}}{\partial s} \right], \quad (\text{S2.60})$$

$$N_{ijk}^{(2,4,0)} = 0, \quad (\text{S2.61})$$

$$N_{ijk}^{(2,4,1)} = \frac{8\gamma_c A}{735\eta D^2} a_{2,0}^{(2)} \overline{u_i S_{jk}} \frac{\partial Q_2^{(0)}}{\partial s}. \quad (\text{S2.62})$$

S3. SOME CALCULATIONS INCLUDING THE OSEEN TENSOR

In this section, we summarize useful results for calculation including Oseen tensor. First it has been obtained [1] that

$$\int da n_i(a') \mathbb{T}_{ij}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) Y_l^m(a) = E_l Y_l^m(a') \quad (\text{S3.1})$$

with

$$E_l = \frac{R}{\eta} \frac{2l(l+1)}{(2l-1)(2l+1)(2l+3)}. \quad (\text{S3.2})$$

We may also have [2]

$$\int da \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_i(a) n_k(a) = \frac{R_0}{5\eta} \delta_{ij} + \frac{R_0}{15\eta} n_i(a') n_j(a') \quad (\text{S3.3})$$

$$\int da \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_i(a) n_k(a) [\delta_{jl} - n_j(a') n_l(a')] = \frac{R_0}{5\eta} [\delta_{il} - n_i(a') n_l(a')]. \quad (\text{S3.4})$$

We may also use the following integral

$$\int \mathbb{T}_{ij}(\mathbf{r}(a), \mathbf{r}(a')) da = \frac{2R_0}{3\eta} \delta_{ij}. \quad (\text{S3.5})$$

In the main text we encounter the integral $\int n_i(a) \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) \nabla_{s,j} Y_l^m(a) da$. Since this integral is difficult to evaluate, we instead consider the following integral

$$\int [n_i(a') \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) n_{\alpha_1}(a) n_{\alpha_2}(a) \cdots n_{\alpha_m}(a)] da. \quad (\text{S3.6})$$

Using integration by part, above integral with a gradient operator is replaced by (S3.6). Let us summarize the results for $m = 1, 2, 3$. For $m = 1$, (S3.6) is readily obtained using (S3.3)

$$\int [n_i(a') \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) n_\alpha(a)] da = \frac{R_0}{15\eta} n_i(a') [3\delta_{k\alpha} + n_k(a') n_\alpha(a')]. \quad (\text{S3.7})$$

For $m = 2$, we have

$$\begin{aligned} & \int [n_\alpha(a) \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) n_\beta(a)] da \\ &= \frac{2R_0}{35\eta} n_\alpha(a') n_\beta(a') n_k(a') - \frac{4R_0}{105\eta} \delta_{\alpha\beta} n_k(a') + \frac{R_0}{35\eta} (\delta_{\alpha k} n_\beta(a') + \delta_{\beta k} n_\alpha(a')), \end{aligned} \quad (\text{S3.8})$$

and for $m = 3$

$$\begin{aligned}
& \int [\mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a'))n_j(a)n_\alpha(a)n_\beta(a)n_\gamma(a)] da \\
&= \frac{R_0}{21\eta} n_\alpha(a')n_\beta(a')n_\gamma(a')n_k(a') + \frac{R_0}{105\eta} [\delta_{\alpha k}n_\beta(a')n_\gamma(a') + \delta_{\beta k}n_\alpha(a')n_\gamma(a') + \delta_{\gamma k}n_\alpha(a')n_\beta(a')] \\
&+ \frac{4R_0}{105\eta} [\delta_{\alpha\beta}\delta_{\beta k} + \delta_{\alpha\gamma}\delta_{\beta k} + \delta_{\alpha k}\delta_{\beta\gamma}].
\end{aligned} \tag{S3.9}$$

In order to obtain the above results, we first calculate the integral

$$\int da n_i(a) \mathbb{T}_{ij}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) Y_l^m(a). \tag{S3.10}$$

We follow the method in [1] in which the similar integral was calculated. This integral is rewritten using the Oseen tensor in Fourier space. Here we consider more general relation for using $\mathbf{n}^{(d)}$ rather than \mathbf{n} :

$$\begin{aligned}
& n_i^{(d)}(a) \mathbb{T}_{ij}(\mathbf{r}(a), \mathbf{r}(a')) n_j^{(d)}(a) Y_l^m(a) \\
&= \frac{1}{\eta R^2} \int da \int_{\mathbf{k}} \frac{1}{k^2} R_i(a) R_j(a) \left(\delta_{ij} - \frac{k_i k_j}{k^2} \right) e^{ik_\alpha [R_\alpha(a) - R_\alpha(a')]} Y_l^m(a) \\
&= \frac{1}{\eta R^2} \int da \int_{\mathbf{k}} \frac{1}{k^2} R^2 e^{ik_\alpha [R_\alpha(a) - R_\alpha(a')]} Y_l^m(a) - \frac{1}{\eta R^2} \int da \int_{\mathbf{k}} \frac{1}{k^4} (R_i(a) k_i) (R_j(a) k_j) e^{ik_\alpha [R_\alpha(a) - R_\alpha(a')]} Y_l^m(a)
\end{aligned} \tag{S3.11}$$

The first term in (S3.11) becomes

$$\begin{aligned}
& \frac{1}{\eta} \int da \int_{\mathbf{k}} \frac{1}{k^2} e^{ik_\alpha [R_\alpha(a) - R_\alpha(a')]} Y_l^m(a) \\
&= \frac{1}{\eta} \int da \int_{\mathbf{k}} \frac{16\pi^2}{k^2} \sum_{l', m'} i^{l'} j_{l'}(kR(a)) Y_{l'}^{m'}(\theta_k, \varphi_k) Y_{l'}^{m'*}(\theta, \varphi) \sum_{l'', m''} (-i)^{l''} j_{l''}(kR(a')) Y_{l''}^{m''*}(\theta_k, \varphi_k) Y_{l''}^{m''}(\theta', \varphi') Y_l^m(a) \\
&= \frac{1}{\eta} \int da \frac{2}{\pi} \sum_{l', m'} \int dk j_{l'}(kR(a)) Y_{l'}^{m'*}(\theta, \varphi) j_{l'}(kR(a')) Y_{l'}^{m'}(\theta', \varphi') Y_l^m(a) \\
&= \frac{2}{\pi\eta} \sum_{l', m'} Y_{l'}^{m'}(a') \int da \mathcal{C}_{l'}(a, a') Y_{l'}^{m'*}(a) Y_l^m(a)
\end{aligned} \tag{S3.12}$$

where we have used (37). Here we define

$$\begin{aligned}
\mathcal{C}_l(a, a') &= \int_0^\infty j_l(kR(a)) j_l(kR(a')) dk \\
&\simeq \frac{\pi}{2(2l+1)R_0} \left[1 - \frac{1}{2R_0} (\delta R(a) + \delta R(a')) \right],
\end{aligned} \tag{S3.13}$$

where the spherical Bessel function is expanded as

$$\begin{aligned}
j_l(kR) &\simeq j_l(kR_0) + k\delta R j_l'(kR_0) \\
&= j_l(kR_0) + \frac{1}{2l+1} k\delta R [l j_{l-1}(kR_0) - (l+1) j_{l+1}(kR_0)].
\end{aligned} \tag{S3.14}$$

The second term in (S3.11) is calculated as

$$\begin{aligned}
& -i^2 \frac{1}{\eta R^2} \int da \int_{\mathbf{k}} \frac{1}{k^4} \frac{\partial^2}{\partial b^2} e^{ik_\alpha [R_\alpha(a) - R_\alpha(a')]} Y_l^m(a) \Big|_{b=1} \\
&= \frac{1}{\eta R^2} \int da \int_{\mathbf{k}} \frac{16\pi^2}{k^4} \frac{\partial^2}{\partial b^2} \sum_{l', m'} i^{l'} j_{l'}(bkR(a)) Y_{l'}^{m'}(\theta_k, \varphi_k) Y_{l'}^{m'*}(\theta, \varphi) \sum_{l'', m''} (-i)^{l''} j_{l''}(kR(a')) Y_{l''}^{m''*}(\theta_k, \varphi_k) Y_{l''}^{m''}(\theta', \varphi') Y_l^m(a) \Big|_{b=1} \\
&= \frac{2}{\eta\pi} \sum_{l', m'} Y_{l'}^{m'}(\theta', \varphi') \int da \bar{\mathcal{C}}_{l'}(a, a') Y_{l'}^{m'*}(\theta, \varphi) Y_l^m(a)
\end{aligned} \tag{S3.15}$$

where

$$\begin{aligned}
\bar{C}_l(a, a') &= \int_0^\infty j_l''(kR(a))j_l(kR(a'))dk \\
&= -\frac{R(a')}{R(a)} \int_0^\infty j_l'(kR(a))j_l'(kR(a'))dk \\
&= -\frac{2l(l+1)-1}{(2l-1)(2l+1)(2l+3)} \left(1 + \frac{\delta R(a')}{R_0} - \frac{\delta R(a)}{R_0}\right) \frac{\pi}{2R_0}.
\end{aligned} \tag{S3.16}$$

Combining (S3.13) and (S3.16), we obtain for $\mathbf{n}^{(d)} \simeq \mathbf{n}$ in which no coupling between modes:

$$\int da n_i(a) \mathbb{T}_{ij}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) Y_l^m(a) = \frac{R_0}{\eta} \frac{2(l^2 + l - 1)}{(2l - 1)(2l + 1)(2l + 3)} Y_l^m(a'). \tag{S3.17}$$

Now we outline the calculation of (S3.6). Let us consider for (S3.8)

$$\int [\mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) n_\alpha(a) n_\beta(a)] da. \tag{S3.18}$$

Respecting the symmetry of the integral, that is, invariance under $\alpha \leftrightarrow \beta$, the integral is decomposed into several third-rank tensors as

$$\begin{aligned}
&\int [n_\alpha(a) \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) n_\beta(a)] da \\
&= \mathcal{A}^{(3)} n_\alpha(a') n_\beta(a') n_k(a') + \mathcal{B}_1^{(3)} \delta_{\alpha\beta} n_k(a') + \mathcal{B}_2^{(3)} (\delta_{\alpha k} n_\beta(a') + \delta_{\beta k} n_\alpha(a')).
\end{aligned} \tag{S3.19}$$

First, by multiplying $n_\alpha(a') n_\beta(a') n_k(a')$, we have

$$\int \left[(n_\alpha(a) n_\alpha(a'))^2 \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a) n_k(a') \right] da = \mathcal{A}^{(3)} + \mathcal{B}_1^{(3)} + 2\mathcal{B}_2^{(3)}. \tag{S3.20}$$

The integral is calculated as

$$\mathcal{A}^{(3)} + \mathcal{B}_1^{(3)} + 2\mathcal{B}_2^{(3)} = \frac{R_0}{8\eta} \int_0^\pi \sin \theta \cos^2 \theta \frac{\cos \theta - \sin^2 \theta / 2}{\sin \theta / 2} d\theta = \frac{8R_0}{105\eta}. \tag{S3.21}$$

When $\alpha = \beta$, (S3.19) becomes

$$\int [\mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_j(a)] da = \mathcal{A}^{(3)} n_k(a') + 3\mathcal{B}_1^{(3)} n_k(a') + 2\mathcal{B}_2^{(3)} n_k(a'). \tag{S3.22}$$

The integral must vanish because uniform normal force never generates flow in an incompressible system, and thus $\mathcal{A}^{(3)} + 3\mathcal{B}_1^{(3)} + 2\mathcal{B}_2^{(3)} = 0$. When $\alpha = k$, (S3.19) becomes

$$\int [n_j(a) \mathbb{T}_{jk}(\mathbf{r}(a), \mathbf{r}(a')) n_k(a) n_\beta(a)] da = \mathcal{A}^{(3)} n_\beta(a') + \mathcal{B}_1^{(3)} n_\beta(a') + 4\mathcal{B}_2^{(3)} n_\beta(a'). \tag{S3.23}$$

The integral is calculated with (S3.17) identifying $n_\beta(a')$ as Y_1^m , and we obtain $\mathcal{A}^{(3)} + \mathcal{B}_1^{(3)} + 4\mathcal{B}_2^{(3)} = \frac{2R_0}{15\eta}$. Note that integration of (S3.8) with respect to a' leads to zero for both sides of this equation. The same argument applies to more complicated integral of (S3.9). We may check the result (S3.9) by multiplying R_0/Ω and integrating over a' for both sides using (D2), (D3), and (S3.5). Both sides result in $2R_0/15\eta$.

[1] T. Ohta, Ann. Phys. **158**, 31 (1984), ISSN 0003-4916.

[2] S. Yabunaka, T. Ohta, and N. Yoshinaga, J. Chem. Phys. **136**, 074904 (pages 8) (2012).