Phenomenological model for ordered onions under shear flow

Kenta Odagiri and Kazue Kudo

Abstract

We propose a simple phenomenological model for the ordered and disordered phases of the multi-lamellar vesicles (onions) induced by shear flow. In experiments on a nonionic surfactant ($C_{12}E_4$) system, the order-disorder transition accompanied with a size jump has observed in the onion phases under a fixed shear flow within a certain range by changing temperature. Our model can simulate ordered and disordered onion phases with different onion sizes. We show numerical results of the onion formation simulated by the model and also discuss what factors in this system are critical to cause the transition between these two different onion phases.

1   Introduction

Surfactants in water form various kinds of assemblies such as lamellar, micelles and vesicles. Multi-lamellar vesicles, which are also called onions, are one of the interesting morphologies induced by shear flow. The onion formation under shear flow has been, in fact, observed in experiments [1, 2, 3, 4]. Onions fill space with various sizes and forms of polyhedra. Each onion consists of concentric lamellar membranes, which were observed by electro-microscopy [5]. The size of an onion depends on the shear rate and the characteristics of a surfactant membrane. Several theories have been proposed to estimate onion size. Diat *et al.* proposed the model in which applied shear stress is balanced by the elastic stress given by the curvature energy of membrane [1]. On the other hand, van der Linden *et al.* proposed the idea that applied shear stress is balanced by the energy cost to deform an onion [6]. These models have been examined by experiments especially concerning shear-rate dependence. The formation of onions, i.e. closed-packed or disordered structure, is another problem to be investigated. Panizza *et al.* suggested that the disordered onion phase is made of randomly oriented monodomains which consist of closed-packed onions [7].

The characteristics of a surfactant membrane, e.g. elasticity, depend on temperature as well as substances. An interesting behavior related to the temperature dependence has been observed in the shear-induced ordering of onions in a nonionic surfactant ($C_{12}E_4$) system: Onions form a two-dimensional (2D) close-packed honeycomb structure in a certain temperature interval and they are disordered below and above the temperature interval [8, 9]. Moreover, in recent experiments, it has been revealed that the order-disorder transition is accompanied by a size jump [9]. The size of onions is 5–6 times larger in the ordered phase than in the disordered one. To our knowledge, no theoretical model about the transition has been proposed, though the order-disorder transition with a size jump had already been reported in other systems [2, 10].

In this paper, we propose a model to simulate ordered and disordered onion phases with different onion sizes from the view point of pattern formation. The model provides the means to demonstrate the formation of an onion phase in real space. Experiments by the small-angle light scattering (SALS) and small-angle X-ray scattering (SAXS) give only images in reciprocal space. It is difficult in those scattering experiments to see what structure onions form in three dimensions. Simulations in real space will give a clue to analyze the structure of the onion phase which is observed experimentally. In this paper, we focus on the onion formation induced by shear flow. Our model illustrates the onion pattern which is stable under shear flow, although the model is not suitable to discuss the lamellar-onion transition.

From the view point of pattern formation, 2D hexagonal structure such as the ordered onion phase appears ubiquitously. One of the most famous examples is nonequilibrium fluid dynamics known as Rayleigh-Bénard convection, of which pattern formation is well described by the Swift-Hohenberg (SH) equation. The SH equation is a well-known model equation for nonequilibrium pattern formation phenomena [11], and it has a property of so-called potential dynamics. Our focus is on the derivation of a simple phenomenological model, which is similar to the SH equation, of the onion formation by constructing the free energy of onion phases under shear flow.
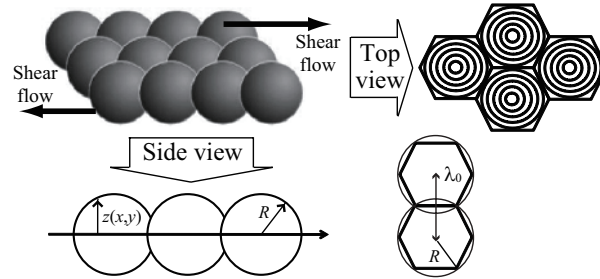
Figure 1: Schematic representation of the shear-induced onion phase in our model.
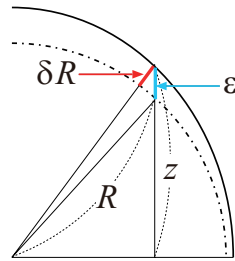
Figure 2: Schematic representation of a distorted vesicle surface. Solid and dot-dash curves denote the surface of a distorted vesicle and that of a spherical one, respectively.

The rest of this paper is organized as follows. In Sec. 2, we introduce a phenomenological model describing the onion formation induced by shear flow. In Sec. 3, we show numerical results of ordered and disordered onion phases with different onion sizes. Section 4 is devoted to the discussion about how the onion size and the ordering structure of onions are selected. Finally, we conclude this paper in Sec. 5.

2   Model

We here construct a phenomenological model for the shear-induced onion phase. We mainly focus on the transition of the onion formation (size and ordering structure) and do not consider the transition between lamellar and onion phases in our model. To simulate the onion formation under shear flow, we derive the free energy of onion phases including the contribution from shear flow, so that we can realize the stationary distribution of onions by minimizing the free energy.

We first assume that onions, which have a certain identical size R, are packed to form a 2D board. The centers of onions are placed on the x-y plane. The spatial distribution of onions is expressed as the distribution of the height of onion surface, $z(x, y)$ (see Fig. 1).

Second, considering onions are closed-packed to form a 2D honeycomb structure (Fig. 1), we assume that the shape of an onion is a slightly-distorted sphere (see Fig. 2) because spherical onions cannot fill space without deformations. In fact, actual onions show a polyhedral structure in the experimental observation [5], and the distortion of the membranes is localized at corners. However, we assume the distortion of membranes is uniformly distributed over slightly-distorted spherical vesicles for simplicity. The validity of this assumption is discussed in the Sec. 4.

2.1   Free energy for the onion phase under shear flow

On the above assumptions, we give the following Gibbs free energy G including the contribution from shear flow.

$$G = F_{\text{onion}} + F_{\text{surface}} - \sigma A. \tag{1}$$

The free energy G, which is a function of $z(x, y)$, consists of two parts: (i) the contribution from the free energy of a deformed onion (F_{onion}) and (ii) the contribution from the stress applied to the surface membrane of packed onions under shear flow ($F_{\text{surface}} - \sigma A$), where F_{surface} is the bending free energy of the onion surface, and σ and A denote the effective stress and the total area of the membrane, respectively. The effective stress σ comes from not only surface tension but also shear stress. The detail of σ is explained later. We derive these two parts of the free energy in what follows.

2.1.1   Contribution from a deformed onion

We first derive the bending free energy of a deformed onion F_{onion} in a similar manner to that of Ref. [12]. The bending free energy per area F/A for a bilayer membrane is represented as the following function of the mean and Gaussian curvatures, H and $\bar{H}$, respectively.

$$\frac{F}{A} = 2\kappa H^2 + \bar{\kappa}\bar{H} + \frac{1}{4}c_1 H^4 + \frac{1}{4}c_2\bar{H}^2 + 2c_3 H^2\bar{H}, \tag{2}$$

where κ and $\bar{\kappa}$ are the conventional bending and Gaussian curvature moduli, respectively, c_1, c_2 and c_3 are nonlinear moduli. We here consider a vesicle which has a spherical shape, which indicates that H^2 and $\bar{H}$ are equivalent. Thus, the free energy, $F_{\text{shell}}(r)$, of a spherical shell with radius r is given by

$$F_{\text{shell}}(r) = 4\pi\tilde{\kappa}H^2 r^2 + \pi\tilde{c}H^4 r^2, \tag{3}$$

where $\tilde{\kappa} \equiv 2\kappa + \bar{\kappa}$ and $\tilde{c} \equiv c_1 + c_2 + 8c_3$.

We next consider distortion of the spherical shell due to packed-onion formation. We assume that the radius of curvature changes from an ideal radius r to that of a distorted spherical shell $r + \delta r$ (Fig. 2). Since the degree of distortion is quite smaller than the ideal radius ($r \gg \delta r$), we expand the mean curvature H in $\alpha = \delta r/r$ and keep terms up to the third order.

$$H = \frac{1}{r + \delta r} \simeq \frac{1}{r}(1 - \alpha + \alpha^2 - \alpha^3). \tag{4}$$

Similarly, we have H^2 and H^4 as follows.

$$H^2 = \frac{1}{r^2}(1 - 2\alpha + 3\alpha^2 - 4\alpha^3), \tag{5}$$

$$H^4 = \frac{1}{r^4}(1 - 4\alpha + 10\alpha^2 - 20\alpha^3). \tag{6}$$

Substituting these expressions into eq. (3), we have

$$F_{\text{shell}}(r) = 4\pi\tilde{\kappa}(1 - 2\alpha + 3\alpha^2 - 4\alpha^3) + \frac{\pi\tilde{c}}{r^2}(1 - 4\alpha + 10\alpha^2 - 20\alpha^3). \tag{7}$$

Thus, the free energy $f_{\text{onion}}(R)$ (per unit volume of an onion with radius R made of n vesicles with layer spacing d) is given by

$$f_{\text{onion}}(R = nd) = \sum_{j=1}^{n} \frac{F_{\text{shell}}(r = jd)}{\frac{4}{3}\pi R^3}$$

$$= \frac{3\tilde{\kappa}}{dR^2}(1 - 2\bar{\alpha} + 3\bar{\alpha}^2 - 4\bar{\alpha}^3) + \frac{3\tilde{c}}{4d^2 R^3}\left(1 - \frac{d}{R}\right)(1 - 4\bar{\alpha} + 10\bar{\alpha}^2 - 20\bar{\alpha}^3), \tag{8}$$

where $\bar{\alpha} = \delta R/R$ is the ratio of distortion on the outermost shell of an onion. Let us consider here the distortion of the onion surface. As illustrated in Fig. 2, the difference between the height of the distorted outermost shell and that of the spherical one is taken to be ϵ, which leads the following relation: $R^2 - (z - \epsilon)^2 = (R + \delta R)^2 - z^2$. Under the conditions of slight distortion ($R \gg \delta R$ and $z \gg \epsilon$), we obtain the following relation

$$\bar{\alpha} = \delta R/R = (\epsilon/R^2)z \equiv \tilde{\alpha}z. \tag{9}$$

By using the above relation, we finally obtain the free energy of a deformed onion F_{onion} as a function of $z(x, y)$,

$$
\begin{aligned}
F_{\mathrm{onion}} &= \int dx dy \Big[f_{\mathrm{onion}}(R) \cdot z \Big] \\
&= \int dx dy \Big[\frac{3\tilde{\kappa}}{dR^2} \big(-(4 + 20\lambda)\tilde{\alpha}^3 z^4 + (3 + 10\lambda)\tilde{\alpha}^2 z^3 - (2 + 4\lambda)\tilde{\alpha} z^2 + (1 + \lambda)z \big) \Big],
\end{aligned}
\tag{10}
$$

where $\lambda = \frac{(R - d)\tilde{c}}{4dR^2\tilde{\kappa}}$.

2.1.2   Contribution from the stress applied to the surface membrane under shear flow

We next consider the free energy from the stress applied to the surface membrane of packed onions under shear flow. Suppose that onion surfaces are covered by a single membrane as if the membrane is a part of each onion surface, and also that the membrane is subjected to an effective stress that comes from surface tension and shear stress. From these assumptions, the free energy contribution from the effective stress is written as

$$
F_{\mathrm{surface}} - \sigma A = \int dA (2\kappa H^2 - \sigma),
\tag{11}
$$

where σ and A denote the effective stress and the total area of the membrane, respectively. The first term is the bending free energy of the onion surface as a single membrane and the second term is the effective stress applied to the membrane under shear flow. We assume the effective stress σ includes contributions not only from effective surface tension but also from shear stress. Actually, since there is no method to distinguish between the contribution from effective surface tension and the contribution from shear stress in the shear-induced onion formation, the effective stress σ may be an indefinite parameter. Nevertheless, we consider σ as one of important factors to form the onion phase. Although the shear can affect vesicle membranes inside onions, we impose the effect from shear stress on only the surface membrane for simplicity.

Using $\boldsymbol{s} = \nabla z$, we have the following expressions [13]

$$
dA = \sqrt{1 + \boldsymbol{s}^2} dx dy,
\tag{12}
$$

$$
H = \frac{\nabla \cdot \boldsymbol{s}}{\sqrt{1 + \boldsymbol{s}^2}}.
\tag{13}
$$

Substituting these expressions into eq. (11) and assuming $\boldsymbol{s}^2 \ll 1$, we finally obtain

$$
F_{\mathrm{surface}} - \sigma A = \int dx dy \Big[2\kappa (\nabla^2 z)^2 - \frac{\sigma}{2} (\nabla z)^2 - \sigma \Big].
\tag{14}
$$

2.2   Distribution of the onion surface $z(x, y)$

To simulate the stationary distribution of the onion surface $z(x, y)$, we use the following Ginzburg-Landau (GL) equation [14], in which time evolution is given by the variation of the free energy G,

$$
\frac{\partial z}{\partial t} = -L \frac{\delta G}{\delta z} = -\tilde{f}_{\mathrm{onion}}(z) + \sigma \nabla^2 z - 4\kappa \nabla^4 z,
\tag{15}
$$

where

$$
\tilde{f}_{\mathrm{onion}}(z) = \frac{3\tilde{\kappa}}{dR^2} \Big[-(16 + 80\lambda)\tilde{\alpha}^3 z^3 + (9 + 30\lambda)\tilde{\alpha}^2 z^2 - (4 + 8\lambda)\tilde{\alpha} z + (1 + \lambda) \Big].
\tag{16}
$$

Here, the phenomenological coefficient L has the dimension of time over mass and we fix $L = 1$ in the following. By solving the above equation, we finally obtain the stationary distribution of the onion surface $z(x, y)$. Since the stationary distribution is realized by minimizing the free energy G of onion phases under shear flow, we can simulate the onion pattern formation under shear flow by solving eq. (15).

Table 1: System parameters

σ	effective stress	R	radius of an onion
κ	bending modulus	d	layer spacing
$\bar{\kappa}$	saddle-splay modulus	ϵ	shift of z caused
$\tilde{c}$	nonlinear modulus		by distortion

Table 2: System parameters chosen for large (case A) and small (case B) onion phases.

	σ	$\bar{\kappa}$	$\tilde{c}$	d	ϵ
case A	20.0	-13.0	-3.9	0.014	1.20
case B	200.0	-1.0	-3.0	0.013	0.35

It should be noted that eq. (15) has essentially the same form as the SH equation. In other words, the equation we have derived is in the same class as the SH equation, although the mechanism of onion pattern formation is different from that of the system in which the SH equation was originally derived.

In fact, the free energy of a deformed onion F_{onion} consists of bending energy and interaction energy coming from compression of the layers, although we consider only the bending free energy in the above. The lammellar interaction between layers is given by $\frac{B_0}{2}(\frac{\Delta d}{d})^2$, where B_0 is the compression modulus and $\Delta d/d$ is relative variation of layer spacing [2]. We assume that $\frac{\Delta d}{d}$ can be estimated by the relative deformation of the radius of an onion $\frac{\delta R}{R}$, $\frac{\Delta d}{d} \sim \frac{\delta R}{R} = \tilde{\alpha} z$. The interaction energy can be expressed as a correction term of z^3 in eq. (10), and the correction term does not change the mathematical properties of eq. (15). In other words, the correction term is involved in the SH equation. We thus ignore the interaction term for simplicity.

3   Results

We here show numerical results obtained from our model to demonstrate the change between a small-disordered onion phase and a large-ordered one. Table 1 lists the system parameters of the model. The system size is 200×200 (lattice) with the periodic boundary condition. To integrate the model equation, we have used the Crank-Nicolson scheme. Since we are mainly interested in the packed-onion ordering, we focus only on the stationary distribution of the surface height $z(x, y)$ after a sufficiently long time. In our numerical simulations, κ is fixed ($\kappa = 10.0$), because it is considered to be almost constant under experimental conditions for onion phases [15]. R is set by using following relation: $R = 4\pi\sqrt{2\kappa/3\sigma}$. The validity of the relation is discussed in the next section. Two different sets of the other parameters (cases A and B) are listed in Table 2.

Figure 3 shows the distributions of $z(x, y)$ and spatial power spectra for case A and case B. For case A, the size of onions is large ($R \simeq 7.3$) and onions show a honeycomb structure (panel (a)). By contrast, for case B, the size of onions is quite small ($R \simeq 2.3$), although the distribution of onions looks an almost perfect honeycomb structure (panel (b)). However, spatial power spectra for these cases are totally different. Panel (c) shows a perfect six-fold symmetry for case A, although panel (d) shows an isotropic circular spectrum for case B. This isotropic ring indicates that the onion phase consists of some small domains of packed onions (i.e. onion texture [7, 16]): Each small domain has the perfect six-fold symmetry but has different directions. These random directions of small domains result in the isotropic ring spectrum.

Furthermore, the time duration before the distribution of $z(x, y)$ falls into a stationary state is totally different for cases A and B. While the ordered onion phase gradually appears in case A, some small domains appear very quickly in case B. The difference can be explained in terms of the linear-growth rate, which is estimated from the linear stability analysis. Since the linear-growth rate is a very small positive
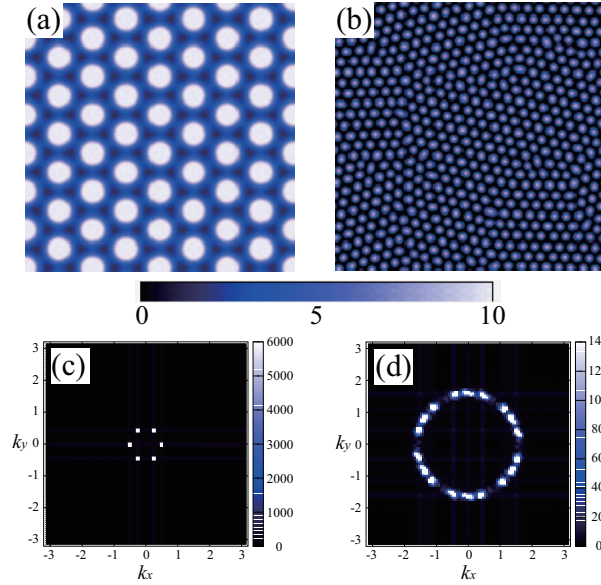
Figure 3: (Upper panels) Spatial distributions of the surface height $z(x, y)$ for (a) case A, and (b) case B. The brighter the color, the larger the value of z. (Lower panels) Spatial power spectra for (c) case A and (d) case B.

value in case A, the stationary pattern can reach the potential minimum, where the ordered honeycomb structure is stable. On the other hand, in case B, because the linear-growth rate is a relatively large positive value, the stationary pattern quickly reaches a metastable state, where some randomly-directed monodomains coexist, and stays there.

We have also checked the robustness of these results by simulations in a larger system size. These results indicate that our model is able to reproduce the change between the large-ordered onion phase and the small-disordered one by changing the system parameters. We next discuss the validity of changing these parameters.

4   Discussion

We here discuss what factors in our model are essential to induce the change between the large-ordered onion phase and the small-disordered one, focusing on the validity and temperature dependence of the parameters in our model.

4.1   Onion size

At first, we focus on how to determine onion size R in our model. As mentioned above, eq. (15) has a similar mathematical structure to that of the SH equation, which is a model equation to simulate periodic domain patterns. The characteristic length λ_0 of the domain pattern produced by the SH equation can be estimated effectively from the linear stability analysis. In the case of eq. (15), $\lambda_0 = 2\pi\sqrt{8\kappa/\sigma}$. λ_0 represents the distance between the centers of neighboring onions and it also has a relation $\lambda_0 = \sqrt{3}R$ because of the geometrical structure of packed onions (see Fig. 1). Therefore, the estimated onion size is given by

$$R = \frac{\lambda_0}{\sqrt{3}} = 4\pi\sqrt{\frac{2\kappa}{3\sigma}}. \tag{17}$$

This equation indicates that the onion size in our model is determined by the ratio of κ and σ. We set R in our simulations by using eq. (17).
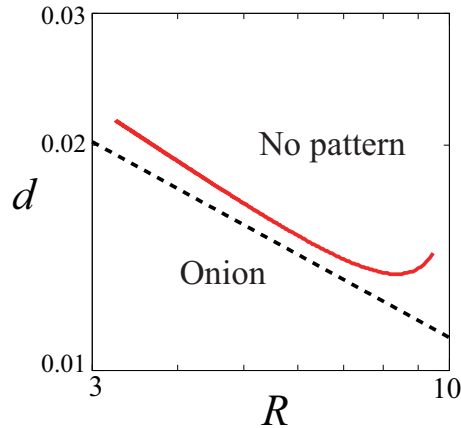
Figure 4: Phase diagram of onion phase as a function of onion size R and layer spacing d. The solid curve indicates the boundary between the onion phase ("Onion") and a phase where no pattern appears ("No pattern"), which is calculated from the linear stability analysis. The slope of the dashed line is $-\frac{1}{2}$.

Actually, only the effective stress σ that comes from surface tension and shear flow is the essential parameter to determine the onion size, because κ is fixed in our simulations. Under a fixed shear rate, it is assumed that the change of σ is caused by only the change of the surface tension. According to experimental studies [7, 17], the elastic modulus G' measured in linear viscoelastic experiments is associated with the surface tension σ_{s} and onion size R: $G' \sim \sigma_{\mathrm{s}}/R$. Assuming $\sigma_s \simeq \sigma$ and substituting eq. (17) into the above relation, we have $G' \sim \sigma_s^{\frac{3}{2}}$. In the experiment of the $C_{12}E_4$ system, on the other hand, G' decreases as the radius of onions increases [15]. These experimental results indicate that σ_s in the large-ordered onion phase is smaller than that in the small-disordered one. This property of the surface tension σ_s corresponds qualitatively to that of the parameter σ in our simulations ($\sigma = 20.0$ and 200.0 in cases A and B, respectively).

We further discuss the change in onion size as a function of shear rate $\dot{\gamma}$ and layer spacing d. Diat et $al.$ estimated onion size R by considering a balance between the elastic stress and the viscous stress as follows [1].

$$R \simeq \sqrt{\frac{4\pi\bar{\kappa}}{\eta d \dot{\gamma}}}, \tag{18}$$

where η is the solvent viscosity. For a fixed η (i.e. Newtonian fluid), this relation implies $R \sim (d\dot{\gamma})^{-\frac{1}{2}}$. This estimation accords with the $\dot{\gamma}^{-\frac{1}{2}}$ dependence which was experimentally observed by themselves. Assuming the effective stress σ in our model is estimated as $\sigma \sim \eta\dot{\gamma}$ and substituting it into eq. (17), we also have $R \sim \dot{\gamma}^{-\frac{1}{2}}$ for a fixed η. We further estimate the relation between onion size R and layer spacing d from the linear stability analysis of eq. (15). Figure 4 shows a phase diagram of onion phase as a function of R and d. The boundary of onion phase, which is calculated from the linear stability analysis, has a slope $-\frac{1}{2}$. It indicates the relation $R \sim d^{-\frac{1}{2}}$, because the ordered onion phase appear near the boundary, that is, the region where the linear-growth rate is very small.

Actually, the order-disorder transition with a size jump is related to the compression of the onion layers. It causes a discontinuous change of the viscosity [18]. The experimental result of Ref. [18] also shows the shear thinning, that is, the viscosity η depends on the shear rate $\dot{\gamma}$. However, the effective stress σ in our model includes not only the shear stress but also η, $\dot{\gamma}$, and d. Thus, we cannot discuss the compression or shear thinning in our model.

4.2   Degree of the distortion

We next consider the validity of the distortion of vesicle membranes due to packing of onions. Although we assume that membranes of packed onions are slightly distorted from a sphere, actual onions have a polyhedral structure [5]. We clarify the validity of the assumption by estimating the degree of the distortion ϵ/R. Inside an onion formed by polyhedral vesicles, the effective surface tension σ_{eff} and the penetration length ξ, which corresponds to the distortion of vesicle membranes, can be estimated as $\sigma_{\text{eff}} \simeq \sqrt{KB}$ and $\xi \sim \sqrt{K/B}$, respectively, where K is the bending modulus $K = \kappa/d$ and B is the compression modulus [7, 19]. We here assume that the effective surface tension corresponds approximately to the effective stress σ in our model, namely $\sigma_{\text{eff}} \simeq \sigma$. We thus have $\xi \sim \kappa/\sigma d$.

On the other hand, the onion size R can be estimated to be $R \sim \sqrt{\kappa/\sigma}$ as mentioned above. We thus obtain the following relation: $\xi/R \sim R$. This indicates that the larger the onion size R the larger the distortion ϵ, if one assumes $\xi \sim \epsilon$. The above relation is also equivalent to the relation $\xi/R^2 = $ constant. This corresponds to our assumption $\tilde{\alpha} = \epsilon/R^2 = $ constant. Therefore, the assumption for the distortion of vesicle membranes due to packing of onions is consistent with other assumptions. In addition, we set ϵ in our simulations to satisfy the above relation that the larger onion has larger distortion.

4.3   Elastic properties of vesicle membranes in packed onions

Elastic properties of vesicle membranes in packed onions, such as κ, $\bar{\kappa}$, and $\tilde{c}$, should also be considered. We first focus on the signs of coefficients of the bending free energy such as κ and $\bar{\kappa}$. In general, the saddle-splay modulus $\bar{\kappa}$ has a negative value unless the sponge (L_3) phase is stable, and an isolated spherical vesicle is stabilized for $\tilde{\kappa} = 2\kappa + \bar{\kappa} < 0$. However, the effects of shear flow are not taken into account in this condition. Thus, it is not necessary in our model to satisfy the condition. We have actually checked simulations for different sets of parameters κ, $\bar{\kappa}$, and $\tilde{c}$ and consequently found that the onion phase can be obtained only when $\bar{\kappa} < 0$ and $\tilde{\kappa} > 0$. This result suggests that the shear stress stabilizes unstable spherical vesicles for $\tilde{\kappa} > 0$.

We further refer two important nonlinear effects of the bending free energy to simulate onion phases in our model: nonlinear modulus in the bending energy and the distortion of membranes due to packing of onions. The usual expression of the Helfrich curvature free energy does not include the nonlinear coefficient $\tilde{c}$. However, $\tilde{c}$ should be considered in an onion phase, because high curvatures are expected near the core of an onion [12]. Also in our model, $\tilde{c}$ is one of the essential parameters to simulate onion phases. Actually, no onion phase is generated from our model if $\tilde{c}$ is taken to be 0. With regard to the distortion of membranes, it is also necessary to consider the distortion of vesicle membranes in our model from the mathematical point of view. The third-order expansion of the curvature on a distorted vesicle leads to the non-linear terms of z in eq. (16), which play a similar role of a double-well potential.

4.4   Temperature dependence of the parameters

We further consider the relation between the temperature dependence of the system parameters in our simulations and that of the physical quantities in experimental studies. Since σ is the critical parameter to determine the onion size in our simulations, it should have a similar temperature dependence to that of the onion size. In other words, σ should be minimized in a certain temperature interval and have larger values above and below the interval.

On the other hand, Kosaka *et al.* [20] reported that layer spacing d can change around the lamellar-onion transition, and also they discussed temperature dependence of $\bar{\kappa}$. They measured d at different concentrations of the surfactant and showed that the ratio δ/d of d and the thickness of bilayers δ has a temperature dependence. Although they focused on the lamellar-onion transition in their experiments, the temperature dependence of δ/d may be one of the important factors to cause the order-disorder transition in onion phases. We therefore change the value of $\bar{\kappa}$ and d in our simulations, assuming the temperature dependence of these parameters. Actually, they are not essential parameters to determine the onion size, but important parameters to determine the linear-growth rate, that is, to determine whether onion phases appear or not.

5   Conclusion

We have proposed the simple phenomenological model, which has the same form as the SH equation, for the onion pattern formation induced by shear flow. We have derived the free energy of an onion phase by considering contributions from deformed packed onions and from the stress applied to the surface membrane on packed onions under shear flow. Our model includes two important nonlinear effects, that is, nonlinear modulus $\tilde{c}$ in the bending energy and the distortion of membranes due to packing of onions. These nonlinearity are crucial in simulations to reproduce onion phases in our model. Then, we have demonstrated that the two different onion phases, large-ordered one and small-disordered one, can be simulated by adjusting the system parameters in our model. The ordered onion phase has a clean honeycomb structure made of packed onions. The disordered onion phase consists of small domains of ordered packed onions. Each domain has the six-fold symmetry, and domains are laid in random directions. The size of packed onions is determined by the ratio of the bending modulus κ and the effective stress σ. Thus, σ is the main control parameter under the fixed-κ condition, though other parameters, such as $\bar{\kappa}$ and d, are also important for emergence of onion phases. In fact, the system parameters in our model, such as σ, are difficult to compare with the physical quantities which can be measured in experiments. In that sense, our model can be improved, nevertheless, it is useful as a simple model for the shear-induced onion formation.

Acknowledgments

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Kenta Odagiri
Meiji Institute for Advanced Study of Mathematical Science, Meiji University, 1-1-1 Higashimita, Tama-ku, Kawasaki 214-8571, Japan.
E-mail: k_oda@meiji.ac.jp

Kazue Kudo
Department of Information Sciences, Ochanomizu University, 2-1-1 Ohtsuka, Bunkyo-ku, Tokyo 112-8610, Japan
E-mail: kudo.kazue@ocha.ac.jp