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Self-organization and pattern formation
Sheet 5

Exercise 14 – The LSW theory of Ostwald ripening

Coarsening describes the continuous growth of the typical length scale in a system. In the Cahn-Hilliard model, coarsening proceeds by Ostwald ripening: larger droplets grow at the expense of smaller droplets which ultimately collapse and vanish. In this exercise, the goal will be to determine the long-time behavior of this system and derive the coarsening law, that is the time evolution of the typical droplet size.

In 1961, I. Lifshitz and V. Slyozov as well as C. Wagner developed a beautiful mean-field theory to describe the Ostwald-ripening process in an ensemble of sparsely dispersed droplets. Assuming a small supersaturation, the droplet radii are small compared to the separation of the droplets. LSW theory therefore assumes that between the droplets (far away from each single one), the system can be described by a uniform concentration $c_\infty(t) = c_- + \epsilon(t)$ which may vary over time (a reservoir). c_- denotes the bulk equilibrium concentration of the low-density phase and $\epsilon(t)$ the supersaturation. Each single droplet is assumed to only interact with this uniform background concentration. In this case, you learned in the lecture that the radius R_i of the i-th droplet evolves as

$$\partial_t R_i(t) = \frac{D_{\text{eff}}^- c_-}{c_+ - c_-} \frac{1}{R_i(t)} \left(\frac{\epsilon(t)}{c_-} - \frac{\ell_\gamma^-}{R_i(t)} \right). \tag{1}$$

Here, D_{eff}^- describes the effective diffusion constant in the low-density phase, c_+ the bulk equilibrium concentration in the high-density phase and ℓ_γ^- the capillary length in the low-density phase.

Using the system volume V, mass conservation implies for the supersaturation

$$V(\bar{c} - c_-) = \epsilon(t) \left(V - \frac{4}{3}\pi \sum_i R_i^3(t) \right) + (c_+ - c_-)\frac{4}{3}\pi \sum_i R_i^3(t) \approx \epsilon(t) V + (c_+ - c_-)\frac{4}{3}\pi \sum_i R_i^3(t), \tag{2}$$

where $\bar{c} > c_-$ is the average concentration in the system and the approximation holds for sparsely dispersed droplets.

To study the dynamics of the ensemble of droplets, we will follow the LSW treatment and analyse the evolution of the droplet-size distribution $P(R^3, t)$. $P(R^3, t)\mathrm{d}R^3$ gives the number density of droplets with a volume in $\frac{4}{3}\pi[R^3, R^3 + \mathrm{d}R^3]$ in the system at time t.

a) Rescale time to find that the dynamics of R_i is given by:

$$\partial_t R^3 = 3\left(\frac{R}{R_c} - 1\right). \tag{3}$$

How is $R_c(t)$ define and what is his interpretation?

b) Argue that the evolution of $P(R^3, t)$ is given by

$$\partial_t P(R^3, t) = -\partial_{R^3}\left[(\partial_t R^3) P(R^3, t)\right], \tag{4}$$

Hint: Consider the evolution of the individual droplets in the ensemble. What is the change of the number of droplets with sizes R^3 in the interval $[R_0^3, R_0^3 + \delta R^3]$ in an infinitesimal time interval dt? Note that, by assumption, a droplet of radius $R > 0$ cannot suddenly vanish or be created, i.e. there must be a corresponding conservation law and continuity equation.

c) Rewrite the condition on the supersaturation into a condition on $R_c(t)$ using the droplet-size distribution P. You should find

$$\bar{c} - c_- = \frac{\ell_\gamma^- c_-}{R_c(t)} + (c_+ - c_-)\frac{4}{3}\pi \int_0^\infty \mathrm{d}R^3\, R^3 P(R^3, t). \tag{5}$$

d) *(Bonus part)* Nondimensionalize the system by introducing the relative droplet size $z(t) = R(t)^3/R_c(t)^3$ and the new time variable $\tau = \log\left(R_c(t)^3/R_c(0)^3\right)$. Why can we use τ to describe the time? How is the distribution $\phi(z,\tau) = P(R^3(z), t(\tau))$ related to the distribution $P(R^3, t)$? Write down the evolution equation for ϕ in the form

$$\partial_\tau \phi(z,\tau) = -\partial_z\left[v(z,\tau)\phi(z,\tau)\right]. \tag{6}$$

You should find

$$v(z,\tau) = -z + \frac{3}{\partial_t R_c(t)^3}\left(z^{\frac{1}{3}} - 1\right). \tag{7}$$

Determine also the constraint on the total mass eq. 5 in terms of τ, z, ϕ.

e) *(Bonus part)* Let us now pick one droplet from the ensemble that started at an initial relative size $z(0) = y$ and evolved to relative size $z(\tau, y)$. Starting from Eq. (6), what is the equation determining the time evolution of z for this droplet? Use an argument similar to the one to derive Eq. (1). What can you say about droplets of initial relative size $y < y_0(\tau)$ where y_0 fulfils $z(\tau, y_0(\tau)) = 0$?
Denote the initial relative size distribution of the droplets by $\phi_0(y)$. Use the evolution of the droplets $z(\tau, y)$ to express the total mass M of the droplets using ϕ_0 and y_0. The total mass M of the droplets is given by

$$M(\tau) = \frac{4}{3}\pi V R_c(0)^3 \mathrm{e}^\tau \int_0^\infty \mathrm{d}z\, z\phi(z,\tau).$$

f) *(Bonus part)* After sufficiently long time, we expect that the relative droplet-size distribution ϕ is independent of the initial size distribution. So the two free quantities to determine are the evolution of $R_c(t)$ and $\phi(z,\tau)$ (see Eqs. (6),(7)). Argue first about the long-time behavior of $R_c(t)$. Let us define $\gamma(\tau) = 3/\left(\partial_t R_c(t)^3\right)$ to simplify the expression for $v(z,\tau)$. Why can we only have $\gamma(\tau) \to \infty, 0, \text{const.}$ for $\tau \to \infty$? Now we exclude the first two possibilities:

- For the first case argue that the total mass in the droplets would diverge as $\tau \to \infty$.
- For the second case show that the total mass in the droplets vanishes. As $v < 0$ for large enough times, all relative droplet sizes z will become small and we can expand $v(z,\tau) \approx -\gamma(\tau) - z$. Solve for $z(\tau, y)$ (in terms of γ), find $y_0(\tau) = 3t(\tau)/R_c(0)^3$ and estimate that the total droplet mass vanishes ($M \to 0$). This violates the constraint on the supersaturation.

g) *(Bonus part)* Let us denote the asymtotic constant γ^* such that $\gamma(\tau) \to \gamma^*$ as $\tau \to \infty$. Use this to determine the asymptotic coarsening law

$$R_c(t)^3 - R_c(0)^3 = \frac{3}{\gamma^*} t. \tag{8}$$

Asymptotically we thus find the so-called LSW scaling $R_c \sim t^{\frac{1}{3}}$. Remember that we rescaled the time t. Reverting back, we find the same dependence on the parameters as in the lecture.

h) *(Bonus part)* Together with the coarsening law we find that the relative droplet-size distribution ϕ converges towards an asymptotic distribution. Use the mass-conservation (supersaturation) constraint to argue that for $\tau \to \infty$ the total mass of the droplets has to become constant. Make the ansatz $\phi(z,\tau) = N(\tau)\psi(z)$ with normalization chosen as $\int_0^\infty \mathrm{d}z \psi(z) = 1$. Determine $N(\tau)$ and the asymptotic equation determining $\psi(z)$. Given this form, why do we say that asymptotically the droplet-size distribution shows scaling? What is special about the shape of the droplet-size distribution?

i) *(Bonus part)* To fully finish up the treatment, we still need to determine the value of γ^*. For this purpose, consider again the evolution of the single droplets $\partial_\tau z = v(z, \tau \to \infty)$, now in the asymptotic regime where $\gamma(\tau) \approx \gamma^*$. Plot the three distinct cases of the flow for the droplets that arise when tuning γ^*. The flow has a bifurcation at a critical value $\gamma^* = \gamma_c$. Explain why values $\gamma^* \lessgtr \gamma_c$ violate mass conservation asymptotically. Argue that $\gamma(\tau)$ has to approach γ_c from below.
Determine the asymptotic value $\gamma^* = \gamma^c$ and show that the normalized asymptotic distribution is given by

$$\psi(z) = \begin{cases} -\frac{1}{v(z,\infty)} \exp\left[\int_0^z \mathrm{d}z' \, \frac{1}{v(z',\infty)}\right], & z < z_{\mathrm{max}} \\ 0, & z > z_{\mathrm{max}} \end{cases}. \tag{9}$$

Here z_{max} fulfils $v(z_{\mathrm{max}}, \infty) = 0$.
Denoting the exponent by $\Psi(z)$, calculate $\partial_z \Psi$ and use it to find

$$\langle z^{\frac{1}{3}} - 1 \rangle = 0. \tag{10}$$

Use this relation to translate the coarsening law found for $R_c(t)$ into the coarsening law for the average droplet radius $\langle R \rangle(t)$. Plot the normalized distribution of R/R_c.

Exercise 15 – The LSW theory in d spatial dimensions

In the previous exercise, we analyzed the LSW theory in $d = 3$ dimensions. The goal of this exercise is to generalize the previous treatment and extend the result to the case of an arbitrary spatial dimension d. We will also derive the LSW result with a different approach.

As in the previous exercise, we rescale time such that the single-droplet kinetics of the radius obey

$$\dot{R}(R,t) \;=\; \frac{1}{R}\left(\frac{1}{R_c(t)} - \frac{1}{R}\right), \tag{11}$$

where $R_c(t) = \frac{\ell_\gamma^- c_-}{\epsilon(t)}$ is the critical radius, which depends on the supersaturation $\epsilon(t)$. Note that the dynamics of the droplet radius do not depend on dimensionality.

In this exercise, we consider the probability distribution $P(R,t)$ of droplet radii (note that in the previous exercise we instead considered the probability distribution of droplet volumes, $P(R^3,t)$).

a) Write down the time evolution of $P(R,t)$ together with the mass conservation in the system. Show that, at large times, the constraint on the total mass takes the form

$$\int_0^\infty \mathrm{d}R\, R^d P(R,t) = \text{const.} \tag{12}$$

b) Following the previous exercise, we now seek a scaling solution for the probability distribution $P(R,t)$. Argue why the ansatz

$$P(R,t) \;=\; \frac{1}{R_c^\alpha(t)}\,\Phi(x)\,, \qquad x := \frac{R}{R_c(t)}, \tag{13}$$

is appropriate, and find the exponent α that ensures total mass conservation.

c) Insert this ansatz into the continuity equation for $P(R,t)$. By exploiting the separation of variables, find one equation governing the dynamics of $R_c(t)$ and one for the profile $\Phi(x)$. What information is contained in the time-dependent equation for the critical radius?

Show that

$$\frac{d}{dt}\,R_c^3(t) \;=\; \text{const},$$

hence

$$R_c(t) \;\propto\; t^{1/3} \quad\Rightarrow\quad z = \frac{1}{3}.$$

You should find the following equation that governs the shape of $\Phi(x)$:

$$-\Big[(d+1)\Phi + x\Phi'\Big] \;+ \gamma^*\,\partial_x\Big[\Big(\tfrac{1}{x} - \tfrac{1}{x^2}\Big)\Phi\Big] = 0. \tag{14}$$

How is γ^* defined?

d) Now specialize to dimension $d = 3$. Show that this equation can be exactly mapped to the profile equation for $\Psi(z)$ found in the previous exercise, where $z = (R/R_c)^3 = x^3$.

Recall that the equation for $\Psi(z)$ is

$$\Psi(z) \;=\; \partial_z\Big(\left[-z + \delta(z^{1/3} - 1)\right] \Psi(z)\Big). \tag{15}$$

For the solution of this differential equation,see Exercise 14 i).

Exercise 16 – Non-reciprocal Cahn-Hillard equation

Consider a system described by two scalar fields ϕ_1, ϕ_2. Their time evolution is governed by a free-energy $F = \int d^dx[f(\phi_1, \phi_2) + \frac{K}{2}(\nabla\phi_1)^2 + \frac{K}{2}(\nabla\phi_2)^2]$. Recall the two types of gradient dynamics governed by a free energy that were studied in class:

$$\text{Allen-Cahn type: } \partial_t\phi_{1/2} = -\Gamma\frac{\delta F}{\delta\phi_{1/2}}, \tag{16}$$

$$\text{Cahn-Hillard type: } \partial_t\phi_{1/2} = \Gamma\nabla^2\frac{\delta F}{\delta\phi_{1/2}}. \tag{17}$$

a) Which underlying assumption were made for the two dynamics? Write the time evolution of the fields ϕ_1, ϕ_2 explicitly in the two cases.

b) Continue this exercise by assuming a Cahn-Hillard type of time-evolution. We can generalize the equations by introducing a new term proportional to $\alpha(\phi_1, \phi_2)$ in the system:

$$\partial_t\phi_1 = \Gamma\nabla^2\left(\frac{\delta F}{\delta\phi_1} + \alpha(\phi_1, \phi_2)\phi_2\right) \tag{18}$$

$$\partial_t\phi_2 = \Gamma\nabla^2\left(\frac{\delta F}{\delta\phi_2} - \alpha(\phi_1, \phi_2)\phi_1\right) \tag{19}$$

The new term introduces a non-reciprocity in the system, which can be interpreted as follows: assuming $\alpha > 0$, the field ϕ_2 increases the local chemical potential of the field ϕ_1, such that it wants to avoid region of high ϕ_2. The field ϕ_1 decreases the local chemical potential of the field ϕ_2, such that it is attracted towards regions of high ϕ_1. Argue why the non-reciprocity leads to the existence of traveling waves in the system. What type of pattern would you expect in the reciprocal case instead, where the new introduced terms have the same sign?

c) Next, we consider the following form of non-reciprocal term $\alpha(\phi_1, \phi_2) = \alpha_0 - \alpha_1|\phi|^2$ and free energy density $f(\phi_1, \phi_2) = -1/2|\phi|^2 + 1/4|\phi|^4$ where $|\phi|^2 = \phi_1^2 + \phi_2^2$.
What are the minima (ϕ_1^0, ϕ_2^0) of the local free energy $f(\phi_1, \phi_2)$? Perform a linear stability analysis around one of the minima, which is also symmetric $\phi_1^0 = \phi_2^0$ by expanding in small deviation: $\phi_1 = \phi_1^0 + \delta\phi_1$ and $\phi_2 = \phi_2^0 + \delta\phi_2$.
Find the parameter regimes such that the non-reciprocity $\alpha(\phi_1, \phi_2)$ makes the steady state unstable.

d) Equations (18) and (19) can be rewritten equivalently as an equation for a complex field $\phi = \phi_1 + i\phi_2$ with an amplitude $|\phi| = \sqrt{\phi_1^2 + \phi_2^2}$ and a phase $\theta = \tan^{-1}(\frac{\phi_1}{\phi_2})$. Find the time evolution for the complex field ϕ.

Hint: your result should read $\partial_t \phi = \Gamma \nabla^2 \left(-(1 + i\alpha_0)\phi + (1 + i\alpha_1)|\phi|^2\phi - K\nabla^2\phi\right)$.

e) Next, we focus on the existence of traveling waves in the system. Show that the Ansatz $\phi_{\boldsymbol{q}} = \rho_{\boldsymbol{q}} e^{i(\boldsymbol{q}\dot{r} - \omega(\boldsymbol{q})t)}$ can solve the non-reciprocal Cahn-Hillard equation and find the explicit value of $\omega(\boldsymbol{q})$ and $\rho_{\boldsymbol{q}}$. Interpret the reason why $\omega(\boldsymbol{q} \to 0) \to 0$ and discuss meaning of the sign of $\omega(\boldsymbol{q})$.

f) Simulate equations (18)-(19) given on the provided Mathematica template and list the type of patterns you observe. Use the parameter sets provided in Table 1.

α_0	α_1	$\phi_{1,0}$	$\phi_{2,0}$
1	5	0.25	0.25
5	2	0.2	0.2
4	4	0.25	0.25
4	4	0	0

Table 1: Parameter values for the simulations.

Your solutions should be handed in in moodle by **Wednesday, November 26 th 2025, 10 am.**