RIMS Kôkyûroku Bessatsu
B31 (2012), 13–29

Gradient flow structure of mean-field models for micro phase separation

By

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§ 1. Introduction

Homopolymer molecules are the chain of one kind of monomers, which can be made by polymerization. Due to repulsive forces between unlike monomers, say A- and B-monomers, the different homopolymers tend to segregate. This is called phase separation.

On the other hand, diblock copolymer molecules consist of subchains of two different type of monomers. In the case of copolymers, since the subchains are chemically bonded, two polymer chains can be forced to mix on a macroscopic scale. However on a microscopic scale, the two polymer chains still segregate, and micro-domains rich in A- and B-monomers respectively form patterns. This is called micro phase separation. For more physical background on this phenomenon we refer to [3, 10].

Energetically favorable configurations have been characterized in the Ohta–Kawasaki theory [22] by minimizers of an energy functional of the form

$$E_\epsilon(u) = \int_{(0,L)^3} \frac{\epsilon}{2}|\nabla u|^2 + \frac{1}{\epsilon}W(u) + \frac{\sigma}{2}|(-\Delta)^{-1/2}(u-\overline{u})|^2\,dx\,.$$

Here $\Omega = (0,L)^3 \subset \mathbb{R}^3$ is the domain covered by the copolymers and u is the density of one of the two monomers. The function W is a double potential with two global minima at 0 and 1, ϵ a small parameter depending on the size and mobility of monomers, $\overline{u} = \frac{1}{L^3}\int_{(0,L)^3} u\,dx \in (0,1)$ the average density and $\sigma \in \mathbb{R}_+$ is a parameter related to the polymerization index. The first term in the energy prefers large blocks of monomers,

Received March 31, 2011.
2000 Mathematics Subject Classification(s): 35B27, 35R35, 53C44, 82D60:
Key Words: *homogenization, geometric evolution, copolymer*:
Supported by JST.
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the second favors segregated monomers and the third term prefers a uniform state or a very fine mixture. Competition between these terms leads to minimizers of E_ϵ which represent micro-phase separation. Thus, this variational problem of characterizing minimizers of E_ϵ can be considered as a prototype model of periodic pattern formation.

In the limit $\epsilon \to 0$ the energy E_ϵ can be replaced by its sharp interface limit

$$E(G) = \mathcal{H}^2(\partial G) + \frac{\sigma}{2}\int_{(0,L)^3} \left|(-\Delta)^{-1/2}(\chi_G - \rho)\right|^2 dx\,, \tag{1.1}$$

where $G \subset [0,L)^3$ denotes the region covered by, say, A-monomers, χ_G the characteristic function of G, $\rho = \overline{\chi_G} = \frac{|G|}{L^3} \in (0,1)$ the volume fraction, and $\mathcal{H}^2$ denotes two dimensional Hausdorff measure. We observe also on the level of the sharp interface model the competition between phase separation on the large scale, which is preferred by the first term, and fine mixtures that are preferred by the nonlocal term. Indeed,

$$0 = \inf_{G\in M}\int_{(0,L)^3} \left|(-\Delta)^{-1/2}(\chi_G - \rho)\right|^2 dx, \quad M = \{G \subset [0,L)^3\,;\,|G| = \rho L^3\}$$

is not attained on M since its minimizing sequence oscillates more and more rapidly.

Starting with the pioneering work [19], where the Ohta–Kawasaki theory is formulated on a bounded domain as a singularly perturbed problem and the limiting sharp interface problem as $\varepsilon \to 0$ is identified, there has been a bulk of analytical work. Minimizers of the energy functionals have been characterized in [1, 4, 5, 24], the existence/stability of stationary solutions has been investigated in [20, 21, 23, 25] and a time dependent model has been considered in [9, 11].

A natural way to set up a model for the evolution of the copolymer configuration that decreases energy and preserves the average density is to consider the gradient flow of the energy with respect to the H^{-1} norm. For E_ϵ this leads to

$$u_t = \Delta(-\epsilon\Delta u + \tfrac{1}{\epsilon}W'(u)) - \sigma(u - \overline{u}).$$

The sharp interface limit of the evolution equation is the following extension of the Mullins–Sekerka evolution for phase separation in binary alloys [13]. The normal velocity V of the interface $\partial G = \partial G(t)$ satisfies

$$V = [\nabla w \cdot \vec{n}] \qquad \text{on } \partial G, \tag{1.2}$$

where $[\nabla w\cdot\vec{n}]$ denotes the jump of the normal component of the gradient of the potential across the interface. Here $\vec{n}$ denotes the unit outer normal to G and

$$[f] = \lim_{\substack{x\notin G\\ x\to\partial G}} f(x) - \lim_{\substack{x\in G\\ x\to\partial G}} f(x).$$

The chemical potential w is for each time determined via

$$-\Delta w = 0 \qquad \text{in } (0,L)^3 \backslash \partial G, \tag{1.3}$$

$$w = \kappa + \sigma(-\Delta)^{-1}(\chi_G - \rho) \qquad \text{on } \partial G, \tag{1.4}$$

where κ is the mean curvature (the sum of the principal curvatures) of ∂G. We are interested in the case that the volume of $G(t)$ is preserved in time and can thus impose Neumann or periodic boundary conditions for w on $\partial(0,L)^3$. In the following we will consider a periodic setting and hence always require that the potential w and the phase domain G are $(0,L)^3$-periodic. Local well-posedness of this evolution has been established in [8].

The evolution defined by (1.2)-(1.4) has an interpretation as a gradient flow of the energy (1.1) on a Riemannian manifold.

To define a metric tensor, consider the manifold of $(0,L)^3$-periodic subsets of $\mathbb{R}^3$ with fixed volume, that is,

$$\mathcal{M} = \{G \subset \mathbb{R}^3\,;\, G \text{ is } (0,L)^3\text{-periodic},\ |G \cap [0,L)^3| = \mathrm{Vol}\,\},$$

whose tangent space $T_G\mathcal{M}$ at an element $G \in \mathcal{M}$ is described by all kinematically admissible normal velocities of ∂G, that is,

$$T_G\mathcal{M} \;=\; \left\{ V\colon \partial G \to \mathbb{R}\,;\; V \text{ is } (0,L)^3\text{-periodic},\; \int_{\partial G \cap [0,L)^3} V\, dS \;=\; 0 \right\}.$$

The Riemannian structure is given by the following metric tensor on the tangent space:

$$g_G(V^1, V^2) \;=\; \int_{[0,L)^3} \nabla w^1 \cdot \nabla w^2 \, dx, \tag{1.5}$$

where w^α is $(0,L)^3$-periodic and solves

$$\begin{aligned} -\Delta w^\alpha &= 0 && \text{in } \mathbb{R}^3 \backslash \partial G, \\ [\nabla w^\alpha \cdot \vec{n}] &= V^\alpha && \text{on } \partial G \end{aligned}$$

for $V^\alpha \in T_G\mathcal{M}$ $(\alpha = 1, 2)$.

(1.2)-(1.4) can be regarded as the gradient flow of the energy (1.1) with respect to the metric g. In other words, V satisfies

$$g_{G(t)}(V, \tilde{V}) = -\langle DE(G(t)), \tilde{V} \rangle \tag{1.6}$$

for all $\tilde{V} \in T_{G(t)}\mathcal{M}$.

§ 2. Restriction to spherical particles and mean-field models

In the following we are interested in the regime where the fraction of A-monomers is much smaller than the one of B-monomers. In this case the A-phase consists of a set of many small disconnected approximately spherical particles. This has been established in the sense of Γ-convergence for the sharp-interface functional in [6]. For our evolutionary problem it seems hence natural to restrict the evolution (1.2)-(1.4) to spherical particles by restricting the gradient flow to such morphologies.

For that purpose we define the submanifold $\mathcal{N} \subset \mathcal{M}$ of all sets G which are the union of disjoint balls

$$G = \bigcup_i B_{R_i}(X_i), \qquad \mathrm{mod}\ L\mathbb{Z}^3$$

where the centers $\{X_i\}_i$ and the radii $\{R_i\}_i$ are variable. Hence $\mathcal{N}$ can be identified with an open subspace of the hypersurface

$$\{\mathbf{Y} = \{R_i, X_i\}_i\,;\, (R_i, X_i) \in \mathbb{R}_+ \times [0,L)^3, \frac{4\pi}{3}\sum_i R_i^3 = \mathrm{Vol}\,\} \subset \mathbb{R}^{4N},$$

where N is the number and $i = 1, \cdots, N$ an enumeration of the particles with centers in the periodic box $[0,L)^3$. Since the normal velocity V satisfies $V = \frac{dR_i}{dt} + \frac{dX_i}{dt}\cdot\vec{n}$ on $\partial B_{R_i}(X_i)$, the tangent space can be identified with the hyperplane

$$T_{\mathbf{Y}}\mathcal{N} = \Big\{\mathbf{Z} = \sum_i (V_i\frac{\partial}{\partial R_i} + \xi_i\cdot\frac{\partial}{\partial X_i})\,;\, (V_i,\xi_i) \in \mathbb{R}\times\mathbb{R}^3, \sum_i R_i^2 V_i = 0\Big\} \subset \mathbb{R}^{4N},$$

such that V_i describes the rate of change of the radius of particle i and ξ_i the rate of change of its center. We use the abbreviation $\mathbf{Z} = \{V_i, \xi_i\}_i$ for $\mathbf{Z} = \sum_i (V_i\frac{\partial}{\partial R_i} + \xi_i\cdot\frac{\partial}{\partial X_i})$.

It turns out to be notationally convenient to consider the normalized energy

$$E(\mathbf{Y}) = E_{\mathrm{surf}}(\mathbf{Y}) + \sigma E_{\mathrm{nl}}(\mathbf{Y}),$$

where

$$E_{\mathrm{surf}}(\mathbf{Y}) = \frac{1}{2}\mathcal{H}^2(\partial G) = 4\pi\sum_i \frac{R_i^2}{2},$$
$$E_{\mathrm{nl}}(\mathbf{Y}) = \frac{3}{2}\int_{(0,L)^3} |\nabla\mu|^2\,dx$$

with μ being $(0,L)^3$-periodic and solving

$$-\Delta\mu = \chi_G - \overline{\chi_G}.$$

Let w be the $(0,L)^3$-periodic function solving

$$-\Delta w = 0 \qquad \text{in } \mathbb{R}^3\backslash\partial G,$$
$$[\nabla w\cdot\vec{n}] = V_i + \xi_i\cdot\vec{n} \qquad \text{on } \partial B_{R_i}(X_i)$$

for $\mathbf{Z} = \{V_i, \xi_i\}_i \in T_{\mathbf{Y}}\mathcal{N}$.

From now on we consider an arrangement of particles as described above which evolves according to the gradient flow equation. Then we see that w satisfies

$$\frac{1}{4\pi R_i^2}\int_{\partial B_{R_i}(X_i)} \left(w - \tfrac{1}{R_i} - 3\sigma\mu\right) dS = \lambda(t) \tag{2.1}$$

and

$$\int_{\partial B_{R_i}(X_i)} (w - 3\sigma\mu)\vec{n}\, dS = 0 \tag{2.2}$$

for all i such that $R_i > 0$, with a Lagrange parameter $\lambda(t)$ that ensures volume conservation. Equations (2.1) and (2.2) are the analogue of (1.4) in the restricted setting.

We remark that in general one cannot expect that a smooth solution exists globally. In fact, if the initial configuration consists of a collection of nonoverlapping balls, short time existence and uniqueness of a smooth solution can be established as it has been done in [14] for a related case without nonlocal term. If a particle disappears, the evolution is not smooth; however one can extend the solution continuously by just starting again with the new configuration. The evolution cannot be extended further when particles collide.

The leading order asymptotics of the evolution have been identified by formal asymptotics in [7, 12]. If $\mathcal{R}$ denotes the average radius (see (2.5) for a precise definition) then it turns out that on a time scale

$$t_{\mathcal{R}} \sim \mathcal{R}^3$$

migration of particles can be neglected and the evolution of the radii is governed by an extension of the classical LSW growth law for coarsening of droplets. More precisely, in a dilute regime (see below for a precise definition), the radii of particles evolve approximately according to

$$\frac{d}{dt}R_i \sim \frac{1}{R_i^2}\left(\lambda R_i - 1 - \sigma R_i^3\right), \tag{2.3}$$

where $\lambda = \lambda(t)$ is such that the volume fraction of the particles is conserved. In an early stage this means that larger particles grow while smaller ones shrink and disappear. However, the term σR_i^3 which comes from the nonlocal energy prevents indefinite coarsening and leads to a stabilization of the remaining particles around a stable radius. On a time scale

$$t_X \sim \frac{1}{\rho}\mathcal{R}^3,$$

where ρ is the volume fraction of particles, migration of particles sets in and typically leads to the self-organization of particles in lattice structures.

We are interested in the mean-field model on a time span of order $\mathcal{R}^3$. [15] rigorously derive a mean-field model for the evolution of the size distribution of particles in the limit of vanishing volume fraction, and show that particles, if initially well separated, remain separated over the time span we are considering and thus well-posedness is ensured.

To describe the mean-field models for this time regime, we now introduce the relevant scales and parameters. We define the number density $\frac{1}{d^3}$ of particles by

$$d^3 \sum_i 1 = L^3 \tag{2.4}$$

and the average volume $\frac{4\pi}{3}\mathcal{R}^3$ by

$$\sum_i R_i(0)^3 = \mathcal{R}^3 \sum_i 1. \tag{2.5}$$

In what follows we use the abbreviation $\sum_i = \sum_{i:R_i>0}$.

We identify the evolution in the limit of vanishing volume fraction of particles. More precisely, we consider a sequence of systems characterized by the parameter

$$\varepsilon := \left(\frac{\mathcal{R}}{d}\right)^{1/2} \tag{2.6}$$

in the limit $\varepsilon \to 0$.

Note that we define here the initial number density and the initial average volume. During the evolution d and $\mathcal{R}$ typically increase in time; the parameter ε is however preserved during the evolution.

It is well-known that there is, analogous to electrostatics, a crucial intrinsic length scale in the system, the screening length

$$L_{sc} := \left(\frac{d^3}{\mathcal{R}}\right)^{1/2}, \tag{2.7}$$

which describes the effective range of particle interactions.

In the case that $L \ll L_{sc}$, that is in the very dilute case, one finds on the time scale of order $\mathcal{R}^3$ that the number density of particles with radius r, denoted by $\nu(t,r)$ (suitably normalized), satisfies

$$\partial_t \nu + \partial_r\left(\frac{1}{r^2}(\lambda r - 1 - \sigma r^3)\nu\right) = 0 \tag{2.8}$$

with

$$\lambda(t) = \frac{\int_{\mathbb{R}_+} \nu\, dr + \sigma \int_{\mathbb{R}_+} r^3\, \nu\, dr}{\int_{\mathbb{R}_+} r\nu\, dr}. \tag{2.9}$$

We observe that this is just the formulation of (2.3) on the level of a size distribution.

On the other hand, if $L \sim L_{sc}$, one obtains an inhomogeneous extension where λ is not constant in space but is replaced by a slowly varying field, in the following denoted by $\psi(t,x)$. The joint distribution of particle radii and centers $\nu = \nu(t,r,x)$ satisfies approximately

$$\partial_t \nu + \partial_r \left(\frac{1}{r^2} (\psi r - 1 - \sigma r^3) \nu \right) + \nabla_x \cdot (3\varepsilon^6 \, \nu \nabla \psi) = 0 \tag{2.10}$$

where $\psi = \psi(t,x)$ satisfies for each t

$$\nabla \cdot \left[\left(\frac{1}{4\pi} + \varepsilon^6 \int_{\mathbb{R}_+} r^3 \nu(dr) \right) \nabla \psi \right] - \left(\int_{\mathbb{R}_+} r \nu(dr) \right) \psi + \int_{\mathbb{R}_+} \nu(dr) + \sigma \frac{1}{|\Omega|} \int_{\mathbb{R}_+ \times \Omega} r^3 \, d\nu(r,x) = 0 \tag{2.11}$$

in $\Omega = (0,L)^3$ with the periodic boundary condition on $\partial\Omega$. Note

$$\rho = \frac{4\pi}{3} \varepsilon^6.$$

(2.10)-(2.11) is our mean-field model with a migration term. Note that the last term in the left hand side of (2.10) and $\int_{\mathbb{R}_+} r^3 \nu(dr)$ in the first term in the left hand side of (2.11) are of order ε^6.

Taking the limit $\varepsilon \to 0$ we have the following mean-field model without the migration term. That is, ν satisfies

$$\partial_t \nu + \partial_r \left(\frac{1}{r^2} (\psi r - 1 - \sigma r^3) \nu \right) = 0, \tag{2.12}$$

where $\psi = \psi(t,x)$ satisfies for each t

$$\frac{1}{4\pi} \Delta \psi - \left(\int_{\mathbb{R}_+} r \, \nu(dr) \right) \psi + \int_{\mathbb{R}_+} \nu(dr) + \sigma \frac{1}{|\Omega|} \int_{\mathbb{R}_+ \times \Omega} r^3 \, d\nu(r,x) = 0 \quad \text{in } \Omega, \tag{2.13}$$

with periodic boundary conditions on $\partial\Omega$. Note that we can normalize the distribution such that the volume constraint is

$$\frac{1}{|\Omega|} \int_{\mathbb{R}_+ \times \Omega} r^3 \, d\nu(r,x) = 1.$$

[15] rigorously derive the inhomogeneous mean-field model in the homogenization limit as $\varepsilon \to 0$ for the case $L \sim L_{sc}$. The derivation of the dilute limit can be done along the same lines.

§ 3. Gradient flow structure of mean-field models with migration

In this section, we see that the mean-field model (2.10)-(2.11) can be regarded as a gradient flow. We denote the joint distribution of particle centers and radii at a given time t by ν or ν_t. The natural space for ν is the space $\mathcal{P}$ of probability measures on $[0,\infty)\times\mathbb{T}$ ($\mathbb{T}$ denotes the 3 dimensional flat torus), which have compact support, and whose marginal with respect to x has a bounded Lebesgue density. We identify functions on $\mathbb{T}$ with Ω-periodic functions on $\mathbb{R}^3$.

The mean-field model has an interpretation as a gradient flow on $\mathcal{P}$. More precisely it is the gradient flow of the energy functional

$$E(\nu) = 4\pi\int_{\mathbb{R}_+\times\Omega}\left(\frac{\sigma}{5}r^5+\frac{r^2}{2}\right)d\nu(r,x)+\frac{3\sigma}{2}\int_\Omega|\nabla\mu|^2\,dx \tag{3.1}$$

where μ is an Ω-periodic function solving

$$-\Delta\mu = \frac{4\pi}{3}\left(\int_{\mathbb{R}_+}r^3\nu(dr)-1\right) \tag{3.2}$$

and $\int_\Omega \mu\,dx = 0$. Here we consider under the volume fraction constraint

$$\frac{1}{|\Omega|}\int_{\mathbb{R}_+\times\Omega}r^3\,d\nu(r,x)=1. \tag{3.3}$$

To define a metric tensor, we denote by

$$\tilde{\mathcal{Z}} = (\tilde{v},\tilde{\xi})\in T_\nu\mathcal{P}$$

such that

$$\int_{\mathbb{R}_+\times\Omega}r^2\tilde{v}\,d\nu(r,x)=0$$

our tangent vectors along the distributional solution $\nu=\nu_t(r,x)$ to the continuity equation

$$\partial_t\nu+\partial_r(\tilde{v}\nu)+\varepsilon^3\nabla_x\cdot(\tilde{\xi}\nu)=0.$$

For each time t, let $\tilde{u}(x)$ and $\tilde{\varphi}(x)$ be Ω-periodic functions solving

$$\Delta\tilde{u} = 4\pi\int_{\mathbb{R}_+}r^2\tilde{v}(x,r)\nu(dr), \tag{3.4}$$

$$\Delta\tilde{\varphi} = -\frac{4\pi}{3}\varepsilon^3\nabla_x\cdot\int_{\mathbb{R}_+}r^3\tilde{\xi}(x,r)\nu(dr) \tag{3.5}$$

and $\int_\Omega\tilde{u}\,dx=\int_\Omega\tilde{\varphi}\,dx=0$. Also for each time t, let $\mu(x)$ be Ω-periodic function satisfying (3.2) and $\int_\Omega\mu\,dx=0$. Then since

$$\frac{\partial}{\partial t}\int_{\mathbb{R}_+}r^3\,\nu(dr) = 3\int_{\mathbb{R}_+}r^2\tilde{v}\,\nu(dr)-\nabla_x\cdot\int_{\mathbb{R}_+}r^3\tilde{\xi}\,\nu(dr),$$

we see

$$\frac{\partial \mu}{\partial t} = -\tilde{u} - \tilde{\varphi},$$

and hence

$$\begin{aligned}\langle DE(\nu), \tilde{\mathcal{Z}} \rangle &= \frac{d}{dt}E(\nu) \\ &= 4\pi \int_{\mathbb{R}_+ \times \Omega} (\sigma r^4 + r)\tilde{v}\, d\nu(r,x) - 3\sigma \int_\Omega \nabla\mu \cdot \nabla(\tilde{u} + \tilde{\varphi})\, dx \\ &= 4\pi \int_{\mathbb{R}_+ \times \Omega} (\sigma r^4 + r + 3\sigma r^2 \mu)\tilde{v}\, d\nu(r,x) + 4\pi\varepsilon^3\sigma \int_{\mathbb{R}_+ \times \Omega} \nabla\mu \cdot \tilde{\xi} r^3\, d\nu(r,x).\end{aligned}$$

Similarly for $\mathcal{Z} = (v, \xi) \in T_\nu\mathcal{P}$ such that

$$\int_{\mathbb{R}_+ \times \Omega} r^2 v\, d\nu(r,x) = 0, \tag{3.6}$$

let $u = u(x)$ and $\varphi = \varphi(x)$ be Ω-periodic functions satisfying

$$\Delta u = 4\pi \int_{\mathbb{R}_+} r^2 v(x,r)\nu(dr), \tag{3.7}$$

$$\Delta\varphi = -\frac{4\pi}{3}\varepsilon^3 \nabla_x \cdot \int_{\mathbb{R}_+} r^3 \xi(x,r)\nu(dr), \tag{3.8}$$

and $\int_\Omega u\, dx = \int_\Omega \varphi\, dx = 0$. Using these potentials, we define a metric tensor

$$\begin{aligned}g(\mathcal{Z}, \tilde{\mathcal{Z}}) &= 4\pi \int_{\mathbb{R}_+ \times \Omega} r^3 v\tilde{v}\, d\, d\nu(r,x) + \frac{1}{3}\int_{\mathbb{R}_+ \times \Omega} \frac{4\pi}{3} r^3 \xi \cdot \tilde{\xi}\, d\nu(r,x) + \int_\Omega \nabla(u+\varphi)\cdot\nabla(\tilde{u}+\tilde{\varphi})\, dx \\ &= 4\pi \int_{\mathbb{R}_+ \times \Omega} r^3 v\tilde{v}\, d\nu(r,x) + \frac{1}{3}\int_{\mathbb{R}_+ \times \Omega} \frac{4\pi}{3} r^3 \xi \cdot \tilde{\xi}\, d\nu(r,x) - 4\pi \int_{\mathbb{R}_+ \times \Omega} r^2 \tilde{v}(u+\varphi)\, d\nu(r,x) \\ &\quad - \frac{4\pi}{3}\varepsilon^3 \int_{\mathbb{R}_+ \times \Omega} r^3 \nabla(u+\varphi)\cdot\tilde{\xi}\, d\nu(r,x).\end{aligned} \tag{3.9}$$

Then we will see that (2.10)-(2.11) is a gradient flow of (3.1) with respect to this metric g.

In fact, if

$$\mathcal{Z} = (v, \xi) \in T_\nu\mathcal{P}$$

denotes the steepest descent direction, then it holds the gradient flow equation

$$\langle DE(\nu), \tilde{\mathcal{Z}} \rangle + g(\mathcal{Z}, \tilde{\mathcal{Z}}) = 0, \quad \forall \tilde{\mathcal{Z}} \in T_\nu\mathcal{P}.$$

This implies $r^3 v = -\sigma r^4 - r + r^2(\lambda + u + \varphi - 3\sigma\mu)$ with a Lagrange multiplier $\lambda(t)$, that is,

$$v(x,r) = \frac{1}{r^2}\Big\{ -\sigma r^3 - 1 + r(\lambda + u(x) + \varphi(x) - 3\sigma\mu(x)) \Big\} \tag{3.10}$$

and also

$$\xi(x,r) = \xi(x) = 3\varepsilon^3 \nabla(u + \varphi - 3\sigma\mu). \tag{3.11}$$

We need to solve a system of equations (3.6), (3.7), (3.8) (3.10) and (3.11) for $(v, \xi, u, \varphi, \lambda)$. Set

$$\psi(x) := \lambda + u(x) + \varphi(x) - 3\sigma\mu(x).$$

Then it follows from

$$r^2 v(x,r) = -\sigma r^3 - 1 + r\psi(x) \tag{3.12}$$

and

$$\xi(x) = 3\varepsilon^3 \nabla\psi \tag{3.13}$$

that

$$\begin{aligned}
\Delta\psi &= \Delta u + \Delta\varphi - 3\sigma\Delta\mu \\
&= 4\pi \int_{\mathbb{R}_+} (-\sigma r^3 - 1 + r\psi)\,\nu(dr) - \frac{4\pi}{3}\varepsilon^3 \nabla_x \cdot \int_{\mathbb{R}_+} r^3 \xi\,\nu(dr) + 4\pi\sigma \int_{\mathbb{R}_+} r^3\,\nu(dr) - 4\pi\sigma \\
&= -4\pi \int_{\mathbb{R}_+} \nu(dr) + 4\pi\psi(x) \int_{\mathbb{R}_+} r\,\nu(dr) - 4\pi\varepsilon^6 \nabla_x \cdot \left(\nabla\psi \int_{\mathbb{R}_+} r^3\,\nu(dr) \right) - 4\pi\sigma.
\end{aligned}$$

Hence ψ satisfies (2.11), which determines ψ uniquely. Then v and ξ are determined via (3.12) and (3.13). The continuity equation $\partial_t \nu + \partial_r(v\nu) + \varepsilon^3 \nabla_x \cdot (\xi\nu) = 0$ implies (2.10). Since

$$\int_{\mathbb{R}_+\times\Omega} r^2 v\,d\nu(r,x) = -\sigma \int_{\mathbb{R}_+\times\Omega} r^3\,d\nu(r,x) - \int_{\mathbb{R}_+\times\Omega} d\nu(r,x) + \int_{\mathbb{R}_+\times\Omega} r\psi\,d\nu(r,x) = 0, \tag{3.14}$$

u is determined via (3.7). Finally λ is determined via

$$\lambda = \frac{1}{|\Omega|} \int_\Omega \psi\,dx. \tag{3.15}$$

Then we see that the mean-field model (2.10)-(2.11) is the gradient flow of the energy (3.1) with respect to the metric (3.9), in other words, the dynamical system where at each time the velocity is the element of the tangent space in the direction of steepest descent of the energy.

§ 3.1. Steady states

In this subsection, we will see that steady states for the mean-field models with migration terms are of simple form. Here steady state ν are solutions independent of t, and characterized as $\mathcal{Z} = 0$, or $v = \xi = 0$. Then we see that $u = \varphi = 0$ and hence $\psi \equiv$ const. Thus $\mu \equiv$ const. and in fact $\mu \equiv 0$. This implies that $\psi \equiv \lambda$, constant in t, and

$$\int_{\mathbb{R}_+} r^3\,\nu(dr) \equiv 1. \tag{3.16}$$

Then it follows from (2.11) that it

$$\lambda = \frac{\int_{\mathbb{R}_+} \nu(dr) + \sigma \int_{\mathbb{R}_+} r^3\,\nu(dr)}{\int_{\mathbb{R}_+} r\,\nu(dr)}. \tag{3.17}$$

From (3.12), we see $\lambda r - 1 - \sigma r^3 = 0$, ν-a.e. Let $R_\pm(\lambda)$ be the solutions of $\lambda r - 1 - \sigma r^3 = 0$ such that $R_-(\lambda) \leq (2\sigma)^{-1/3} \leq R_+(\lambda)$ for $\lambda \geq \lambda_{\min} := \min_{r>0}(\frac{1}{r} + \sigma r^2)$. Then the support of measure ν is in $\{R_+(\lambda), R_-(\lambda)\} \times \Omega$ and hence ν is of the form $\delta_{R_+(\lambda)} \otimes a(x) + \delta_{R_-(\lambda)} \otimes b(x)$ where $a(x), b(x) \geq 0$. Setting $\theta(x) = \int_{\mathbb{R}_+} \nu(dr)$, it follows from (3.16) that

$$\nu = \delta_{R_+(\lambda)} \otimes \frac{1 - \theta(x)R_-(\lambda)^3}{R_+(\lambda)^3 - R_-(\lambda)^3} + \delta_{R_-(\lambda)} \otimes \frac{\theta(x)R_+(\lambda)^3 - 1}{R_+(\lambda)^3 - R_-(\lambda)^3}$$

with $R_+(\lambda)^{-3} \leq \theta(x) \leq R_-(\lambda)^{-3}$ if $\lambda > \lambda_{\min}$, and $\nu = 2\sigma\delta_{(2\sigma)^{-1/3}} \otimes 1$ if $\lambda = \lambda_{\min}$.

All these ν satisfy (3.17) and so they are steady states. Note that even for fixed λ, $\theta(x)$ is not necessarily determined uniquely. But the distribution of the radii of particles for one steady state concentrate on at most two numbers, which is independent of x.

§ 4. Restricted gradient flow structure of mean-field models without migration

Taking $\rho \to 0$ we get the mean-field model without the migration term (2.12)-(2.13) with the volume constraint (3.3). In this section we will see that this can also be regarded as a gradient flow. Here we consider the same energy functional as (3.1), but consider only *restricted* tangent vectors

$$T'_\nu\mathcal{P} = \{\mathcal{Z} \in T_\nu\mathcal{P}\,;\, \mathcal{Z} = (v, 0)\},$$

namely,

$$\mathcal{Z} = (v, 0),\ \tilde{\mathcal{Z}} = (\tilde{v}, 0) \in T_\nu\mathcal{P}$$

such that

$$\int_{\mathbb{R}_+\times\Omega} r^2 v\,d\nu(r,x) = 0, \quad \int_{\mathbb{R}_+\times\Omega} r^2 \tilde{v}\,d\nu(r,x) = 0.$$

We denote by $\mathcal{Z} = (v, 0)$ the restricted steepest descent direction as before. Let $\tilde{u}$ and u be Ω-periodic functions satisfying

$$\Delta \tilde{u} = 4\pi \int_{\mathbb{R}_+} r^2 \tilde{v}(x,r)\nu(dr),$$
$$\Delta u = 4\pi \int_{\mathbb{R}_+} r^2 v(x,r)\nu(dr),$$

and $\int_\Omega \tilde{u}\,dx = \int_\Omega u\,dx = 0$. Then the derivative of the energy is

$$\begin{aligned}\langle DE(\nu), \tilde{\mathcal{Z}}\rangle &= 4\pi \int_{\mathbb{R}_+\times\Omega} (\sigma r^4 + r)\tilde{v}\,\nu(drdx) - 3\sigma \int_\Omega \nabla\mu\cdot\nabla\tilde{u}\,dx \\ &= 4\pi \int_{\mathbb{R}_+\times\Omega} (\sigma r^4 + r + 3\sigma r^2\mu)\tilde{v}\,\nu(drdx),\end{aligned}$$

and the metric tensor is

$$\begin{aligned}g(\mathcal{Z}, \tilde{\mathcal{Z}}) &= 4\pi \int_{\mathbb{R}_+\times\Omega} r^3 v\tilde{v}\,\nu(drdx) + \int_\Omega \nabla u\cdot\nabla\tilde{u}\,dx \\ &= 4\pi \int_{\mathbb{R}_+\times\Omega} r^3 v\tilde{v}\,\nu(drdx) - 4\pi \int_{\mathbb{R}_+\times\Omega} r^2\tilde{v}u\,\nu(drdx).\end{aligned}$$

Hence the restricted gradient flow equation

$$\langle DE(\nu), \tilde{\mathcal{Z}}\rangle + g(\mathcal{Z}, \tilde{\mathcal{Z}}) = 0 \quad \text{for all } \tilde{\mathcal{Z}} \in T'_\nu\mathcal{P}$$

turns into

$$v = \frac{1}{r^2}(-\sigma r^3 - 1 + r(\lambda + u - 3\sigma\mu))$$

with the Lagrange multiplier λ. Set $\psi := \lambda + u - 3\sigma\mu$, then similarly as in Section 3, we obtain the mean-field model. This means that the velocity is the element of the tangent space in the direction of steepest descent among all restricted directions.

§ 5. The derivation of mean-field models

In this section we review the derivation result obtained in [15]. We will introduce suitably rescaled variables, set up the equation in the rescaled variables, and under the appropriate assumption on our initial particle arrangement, we derive the mean-field models (2.12)-(2.13). Finally we will describe the main ideas of the proof.

We assume from now on that $L = L_{sc}$ and for the ease of presentation, we will rescale the spatial variables by L_{sc} such that $L_{sc} = L = 1$ and hence $d = \varepsilon$, $\mathcal{R} = \varepsilon^3$. We rescale r by $\mathcal{R}$, the time t by $\mathcal{R}^3$, as a consequence the velocities by $\mathcal{R}^{-2}$, in view of

(1.4) the velocity potentials by $\mathcal{R}^{-1}$, the parameter σ by $\mathcal{R}^{-3}$ and finally the potential in the nonlocal energy by $\mathcal{R}^2$. More precisely, we introduce $\hat{R}_i$, $\hat{t}$, $\hat{V}_i$, $\hat{\xi}$, $\hat{w}$, $\hat{\sigma}$ and $\hat{\mu}$ via

$$\begin{aligned} R_i(t) &= \varepsilon^3 \hat{R}_i(\hat{t}), \\ V_i(t) &= \varepsilon^{-6}\hat{V}_i(\hat{t}), & \xi_i(t) &= \varepsilon^{-6}\hat{\xi}_i(\hat{t}), \\ w(t,x) &= \varepsilon^{-3}\hat{w}(\hat{t},x), & \mu(t,x) &= \varepsilon^6\hat{\mu}(\hat{t},x) \\ \sigma &= \varepsilon^{-9}\hat{\sigma}, & t &= \varepsilon^9\hat{t}. \end{aligned}$$

We remark that in the rescaled variables, $\hat{V}_i = \frac{d\hat{R}_i}{dt}$ is the rate of change of $\hat{R}_i$, but $\hat{\xi}_i = \varepsilon^{-3}\frac{d\hat{X}_i}{dt}$ is not equal to the velocity $\frac{dX_i}{dt}$ of the particle center in the same time scale. However, $\hat{\xi}_i$ as defined above, appears naturally in the homogenization of the metric tensor. In the end, over the time scales we are considering, ξ_i and hence also $\frac{dX_i}{dt}$, vanish in the limit.

From now on we only deal with the rescaled quantities and drop the hats in the notation.

In rescaled variables the submanifold $\mathcal{N}^\varepsilon$ is given by

$$\mathcal{N}^\varepsilon = \{\mathbf{Y}^\varepsilon = \{R_i, X_i\}_i \, ; \sum_i \varepsilon^3 R_i^3 = 1\}$$

and the tangent space by

$$T_{\mathbf{Y}^\varepsilon}\mathcal{N}^\varepsilon = \Big\{\tilde{\mathbf{Z}}^\varepsilon = \sum_i (\tilde{V}_i \frac{\partial}{\partial R_i} + \varepsilon^3 \tilde{\xi}_i \cdot \frac{\partial}{\partial X_i}) \, ; \sum_i R_i^2 \tilde{V}_i = 0\Big\}.$$

We use the abbreviation $\tilde{\mathbf{Z}}^\varepsilon = \{\tilde{V}_i, \tilde{\xi}_i\}_i$ for $\tilde{\mathbf{Z}}^\varepsilon = \sum_i(\tilde{V}_i\frac{\partial}{\partial R_i} + \varepsilon^3\tilde{\xi}_i \cdot \frac{\partial}{\partial X_i})$. We regard $\{\tilde{V}_i, \tilde{\xi}_i\}_i$ as the component of a tangent vector $\tilde{\mathbf{Z}}^\varepsilon$ with respect to a basis $\{\frac{\partial}{\partial R_i}, \varepsilon^3\frac{\partial}{\partial X_i}\}_i$. We will always denote by $\mathbf{Z}^\varepsilon = \{V_i, \xi_i\}_i$ the direction of steepest descent. Recall that $V_i = \frac{dR_i}{dt}$, but $\xi_i = \varepsilon^{-3}\frac{dX_i}{dt}$. The notation $\tilde{\mathbf{Z}}^\varepsilon$ will be used for an arbitrary element of the tangent space. Furthermore we use the abbreviation $B_i := B_{\varepsilon^3 R_i}(X_i)$.

We define the energy in rescaled variables as

$$E_\varepsilon(\mathbf{Y}^\varepsilon) = 4\pi \sum_i \varepsilon^3 \frac{R_i^2}{2} + \frac{3\sigma}{2}\int_{(0,1)^3} |\nabla \mu^\varepsilon|^2 \, dx,$$

where $\mu^\varepsilon = \mu^\varepsilon(t,x)$ is $(0,1)^3$-periodic and solves $-\Delta\mu^\varepsilon = \frac{1}{\varepsilon^6}\chi_{\cup_i B_i} - \frac{4\pi}{3}$ and $\int_{(0,1)^3}\mu^\varepsilon \, dx = 0$.

For the steepest descent directions $\mathbf{Z}^\varepsilon = \{V_i, \xi_i\}_i$, we define the potentials w^ε, u^ε, ϕ^ε, where $w^\varepsilon = u^\varepsilon + \phi^\varepsilon$,

$$\begin{aligned} &\int_{(0,1)^3} \nabla u^\varepsilon \cdot \nabla\zeta \, dx + \sum_i \int_{\partial B_i} \frac{1}{\varepsilon^3} V_i \, \zeta \, dS = 0\,, \\ &\int_{(0,1)^3} \nabla \phi^\varepsilon \cdot \nabla\zeta \, dx + \sum_i \int_{\partial B_i} \frac{1}{\varepsilon^3} \xi_i \cdot \vec{n}\, \zeta \, dS = 0 \end{aligned} \tag{5.1}$$

for all $\zeta \in \overset{\circ}{H}{}^1_p$. Here $\overset{\circ}{H}{}^1_p$ is the subspace of H^1_p of functions with mean value zero, where H^1_p is the natural space for potentials of diffusion fields, the space of functions $w = w(x) \in H^1(\mathbb{T})$. Note that the potentials are only determined up to an additive constant. In what follows we fix this constant by requiring that $\int_{(0,1)^3} u^\varepsilon\, dx = \int_{(0,1)^3} \phi^\varepsilon\, dx = 0$.

Equations (2.1) and (2.2) for the direction of steepest descent, turn into

$$\frac{1}{|\partial B_i|}\int_{\partial B_i} (u^\varepsilon + \phi^\varepsilon - 3\sigma\mu^\varepsilon)\, dS = \frac{1}{R_i} + \lambda^\varepsilon(t) \tag{5.2}$$

for some $\lambda^\varepsilon(t) \in \mathbb{R}$ and

$$\int_{\partial B_i} \big(u^\varepsilon + \phi^\varepsilon - 3\sigma\mu^\varepsilon\big)\vec{n}\, dS = 0 \tag{5.3}$$

for all i such that $R_i > 0$. Here and in what follows we abbreviate, with some abuse of notations, for a ball $B_R(X)$ the surface area by $|\partial B_R|$ and its volume by $|B_R|$.

We denote the joint distribution of particle centers and radii at a given time t by $\nu_t^\varepsilon \in (C^0_p)^*$, which is given by

$$\int \zeta\, d\nu_t^\varepsilon \;=\; \sum_i \varepsilon^3 \zeta\left(R_i(t), X_i(t)\right) \quad \text{for } \zeta \in C^0_p\,, \tag{5.4}$$

where C^0_p stands for the space of continuous functions on $\mathbb{R}_+ \times \mathbb{T}$ which have compact support included in $\mathbb{R}_+ \times \mathbb{T}$. We identify functions $\zeta = \zeta(r,x) \in C^0_p$ with functions which are $(0,1)^3$-periodic in x. Note that since $\zeta(r,x) = 0$ for $r = 0$, particles which have vanished do not enter the distribution. Hence the natural space for ν_t^ε is the space $(C^0_p)^*$ of Borel measures on $\mathbb{R}_+ \times \mathbb{T}$, that is, the product of the positive half axis and the torus.

In accordance with the notation in (5.4) we will use in what follows the abbreviation

$$\int \zeta\, d\nu_t := \int_{\mathbb{R}\times(0,1)^3} \zeta(r,x)\, d\nu_t(r,x) \quad \text{for } \zeta \in C^0_p,\ \nu_t \in (C^0_p)^*.$$

Otherwise the domain of integration is specified.

We can now state the main result which informally says that ν_t^ε converges as $\varepsilon \to 0$ to a weak solution of (2.12)-(2.13).

Let $T > 0$ be given and assume some appropriate assumptions on initial particle arrangements. Then there exists a subsequence, again denoted by $\varepsilon \to 0$, and a weakly continuous map $[0,T] \ni t \mapsto \nu_t \in (C^0_p)^*$ with

$$\int \zeta\, d\nu_t^\varepsilon \;\to \int \zeta\, d\nu_t$$

uniformly in $t \in [0,T]$ for all $\zeta \in C^0_p$, and

$$\int r^3\, d\nu_t \;=\; 1$$

for all $t \in [0,T]$. Furthermore, there exists a measurable map $(0,T) \ni t \mapsto \psi(t) \in H^1_p$ such that (2.12) and (2.13) hold in the following weak sense.

$$\frac{d}{dt}\int \zeta\, d\nu_t \;=\; \int \partial_r\zeta\, \frac{1}{r^2}\left(r\,\psi(t,x) - 1 - \sigma r^3\right) d\nu_t \tag{5.5}$$

distributionally on $(0,T)$ for all $\zeta \in C^0_p$ with $\partial_r\zeta \in C^0_p$. Here

$$\int_{(0,1)^3}\left(\frac{1}{4\pi}\nabla\psi(t,x)\cdot\nabla\zeta - \sigma\zeta\right)dx + \int \big(\psi(t,x)\,r - 1\big)\zeta\, d\nu_t = 0 \tag{5.6}$$

for all $\zeta \in H^1_p$ and almost all $t \in (0,T)$.

Moreover the energy functional converges in the following sense.

$$\lim_{\varepsilon\to 0} E(\mathbf{Y}^\varepsilon) = E(\nu_t), \quad \text{uniformly in } t \in [0,T],$$

where

$$E(\nu_t) = 4\pi\int\left(\frac{r^2}{2} + \sigma\frac{r^5}{5}\right)d\nu_t + \frac{3\sigma}{2}\int_{(0,1)^3}|\nabla\mu|^2\,dx.$$

Here

$$\int_{(0,1)^3}\left(\nabla\mu(t,x)\cdot\nabla\zeta + \frac{4\pi}{3}\zeta\right)dx - \frac{4\pi}{3}\int r^3\zeta\, d\nu_t = 0 \tag{5.7}$$

for all $\zeta \in H^1_p$ and almost all $t \in (0,T)$.

The strategy of the proof is as follows. We first derive some simple a-priori estimates, and then homogenize within the variational principle of a gradient flow structure, also known as the Rayleigh principle. This follows the related analysis in [16] for the case $\sigma = 0$. In contrast to [16], since our particles move, we need to show that the particles remain separated over the time span we are considering. We also have to identify corresponding additional terms in the metric tensor. Furthermore, in order to prove the convergence of the differential of the energy, we need to prove that the tightness condition is preserved in time.

Rayleigh principle says that (1.6) can be reformulated as follows: for fixed t the direction of steepest descent v minimizes

$$\tfrac{1}{2}g_{G(t)}(\tilde V,\tilde V) + \langle DE(G(t)),\tilde V\rangle \tag{5.8}$$

under all $\tilde V \in T_{G(t)}\mathcal{M}$. Since we will in general only deal with solutions which are piecewise smooth in time and globally continuous, it is convenient to have (5.8) in the time integrated version, that is v minimizes

$$\int_0^T \beta(t)\Big(\tfrac{1}{2}g_{G(t)}(\tilde V,\tilde V) + \langle DE(G(t)),\tilde V\rangle\Big)\,dt \tag{5.9}$$

where $\beta = \beta(t)$ is an arbitrary nonnegative smooth function.

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