Macroscopic limit of the Becker–Döring equation via gradient flows

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Abstract. This work considers gradient structures for the Becker–Döring equation and its macroscopic limits. The result of Niethammer [16] is extended to proof the convergence not only for solutions of the Becker–Döring equation towards the Lifshitz–Slyozov–Wagner equation of coarsening, but also the convergence of the associated gradient structures. We establish rigorously the gradient structure of the nonlocal coarsening equation and show continuous dependence on the initial data within this framework. Further, we prove that on the considered time scale the small cluster distribution of the Becker–Döring equation follows a quasistationary distribution dictated by the monomer concentration. Both results help to understand the well preparedness of the initial data for the convergence of solutions.

1. Introduction

The Becker–Döring equation. In this work, we are interested in gradient structures for the Becker–Döring equation and its macroscopic limits. The Becker–Döring equation [3] is a model for the coagulation and fragmentation of clusters consisting of identical monomers. The main modeling assumptions is that only monomers are able to coagulate and fragment with other clusters. Moreover, the conservation of the total density of monomers is imposed

$$\varrho(t) := \sum_{l=1}^{\infty} l n_l(t) = \sum_{l=1}^{\infty} l n_l(0) =: \varrho_0 \qquad \text{for all } t > 0, \tag{1.1}$$

where $n_l(t)$ is the density of clusters of size l at time t. The evolution of the densities $n_l(t)$ is given by an countable number of ordinary differential equations of the form

$$\dot{n}_l(t) = J_{l-1}(t) - J_l(t) \qquad l = 2, 3 \dots \tag{1.2}$$

where J_l is the flux from clusters of size l to clusters of size $l+1$. The system (1.2) gets closed with an equation for n_1

$$\dot{n}_1(t) = \sum_{l=1}^{\infty} J_l(t) - J_1(t) =: J_0(t) - J_1(t),$$

which is chosen, such that formally $\frac{\mathrm{d}}{\mathrm{d}t} \sum_{l=1}^{\infty} l n_l = 0$. The fluxes J_l are given by mass-action kinetics, that is the rate of fragmentation is determined by $a_l n_1 n_l$ and the rate of fragmentation is given by $b_{l+1} n_{l+1}$, where a_l and b_{l+1} are rate factors only depending on l. This leads to the constitutive relation

$$J_l(t) = a_l n_1(t) n_l(t) - b_{l+1} n_{l+1}(t), \qquad l = 1, 2, \dots.$$

The detailed balance condition for this system reads $J_l(t) = 0$ for all l. From there, one obtains the following one-parameter family of equilibrium solutions

$$\omega_l(z) := z^l Q_l \qquad Q_1 := 1 \qquad \text{and} \qquad Q_l := \prod_{j=1}^{l-1} \frac{a_j}{b_{j+1}}. \tag{1.3}$$

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To specify the long-time behavior, we introduce the convergence radius of the series $z \mapsto \sum_l l z^l Q_l$ by $z_s \in [0,\infty]$ as well as its value at the convergence radius

$$\varrho_s := \sum_{l=1}^{\infty} l z_s^l Q_l \in [0,\infty]. \tag{1.4}$$

We are interested in the regime, where $z_s \in (0,\infty)$ and $\varrho_s \in (0,\infty)$. We will assume that the rates are explicitly given as follows:

Assumption 1.1 (Rates). *For $\alpha \in [0,1)$, $\gamma \in (0,1)$ and $z_s, q > 0$ define the coagulation and fragmentation rate of a monomer for a cluster of size l by*

$$a_l := l^\alpha \qquad \text{and} \qquad b_l := l^\alpha \big(z_s + q l^{-\gamma}\big).$$

Hereby, the parameter z_s is consistent with its definition as radius of convergence (cf. Lemma 4.1) and ϱ_s as defined in (1.4) is strictly positive and finite under Assumption 1.1.

Then, as investigated by [2] solutions to the Becker–Döring equation with $\varrho_0 \leq \varrho_s$ converge to the equilibrium state $\omega_l(z)$, where $z = z(\varrho_0)$ is given such that $\sum_{l=1}^\infty l z^l Q_l = \varrho_0$ and the convergence takes place in a weighted ℓ^1 space

$$\lim_{t\to\infty} \sum_{l=1}^{\infty} l \, |n_l(t) - \omega_l(z)| = 0$$

However, if $\varrho_0 > \varrho_s$ then

$$\lim_{t\to\infty} n_l(t) = \omega_l(z_s) \qquad \text{for each } l \geq 1.$$

In this case, the excess mass $\varrho_0 - \varrho_s > 0$ vanishes in the limit $t \to \infty$. The interpretation is, that the excess mass is contained in larger and larger clusters as times evolve. These large clusters form a new phase, e.g. liquid droplets formed out of supersaturated vapor. It is the aim of the is work to add some aspect to the understanding of the formation of the new phase.

The crucial ingredient for the above convergence statements is the existence of a Lyapunov functional of the form

$$\mathcal{F}_z(n) := \sum_{l=1}^{\infty} \left(n_l \left(\log \frac{n_l}{\omega_l(z)} - 1 \right) + \omega_l(z) \right), \tag{1.5}$$

where $z > 0$ is a parameter selecting the stationary state. An calculation shows, that it is formally decreasing along solutions to the Becker–Döring equation

$$\frac{\mathrm{d}\mathcal{F}_z(n(t))}{\mathrm{d}t} = -\sum_{l=1}^{\infty} (a_l n_1 n_l - b_{l+1} n_{l+1})(\log a_l n_1 n_l - \log b_{l+1} n_{l+1}) =: -\mathcal{D}(n(t)) \leq 0. \tag{1.6}$$

Hence, the Lyapunov function can be interpreted as a free energy dissipating along the flow. This indicates, that in the long-time the free energy is minimized. By the mass conservation (1.1), we expect the long-time limit to be the solution to the following minimization problem

$$\inf \left\{ \mathcal{F}_z(n) : \sum_{l=1}^{\infty} l n_l = \varrho_0 \right\} = \begin{cases} \mathcal{F}_z(\omega(z)), & \varrho \leq \varrho_s; \\ \mathcal{F}_{z_s}(\omega(z_s)), & \varrho > \varrho_s. \end{cases} \tag{1.7}$$

In the first case the infimum is attained and the parameter $z = z(\varrho_0)$ is chosen such that $\sum_{l=1}^\infty l \omega_l(z) = \varrho_0$. In the second case the infimum is not attained (cf. [2, Theorem 4.4]). Hence, the functional reflects correctly the long-time behavior of the equation. Moreover, the Lyapunov function has the form of a relative entropy and the question arises, whether their exists a gradient structure for the Becker–Döring equation having this relative entropy as driving free energy.

Reversible chemical reactions and gradient flows. To bring the system into the framework of gradient-flows, it is helpful to interpret the Becker–Döring equation as the following system of chemical reactions

$$X_1 + X_{l-1} \underset{b_l}{\overset{a_{l-1}}{\rightleftharpoons}} X_l, \qquad l = 2, 3, \dots \ . \tag{1.8}$$

Hereby, X_l denotes a cluster of size l and the rates for coagulation $\{a_l\}_{l\geq 1}$ and fragmentation $\{b_l\}_{l\geq 2}$ are positive as in Assumption 1.1. In this formulation, we can use the gradient structure as observed by Mielke [14] for chemical reactions under detailed balance condition and it turns out that the Becker–Döring equation is indeed a gradient flow with respect to the Lyapunov function (1.5) under a suitable metric.

The gradient flow formulation allows for a variational characterization initiated by de Giorgi and its collaborators [6] under the name of curves of maximal slope. For any curve $[0,T] \ni t \mapsto n(t)$ satisfying the mass constraint (1.1), one can define a functional of the form

$$\mathcal{J}(n) = \mathcal{F}(n(T)) - \mathcal{F}(n(0)) + \frac{1}{2}\int_0^T \mathcal{D}(n(t))\,\mathrm{d}t + \frac{1}{2}\int_0^T |n'(t)|^2_{n(t)}\,\mathrm{d}t \geq 0, \tag{1.9}$$

where $|n'(t)|_{n(t)}$ is a suitable weighted norm of the time derivative, $\mathcal{D}$ is the dissipation as defined in (1.6) and $\mathcal{F} = \mathcal{F}_z$ from (1.5) with $z = z(\varrho_0)$ as defined after (1.7). The functional $\mathcal{J}$ characterizes solution as it only vanishes on solutions to the Becker–Döring equation. We will use this variational structure to pass to the limit after a suitable rescaling.

The macroscopic limit. The connection between the Becker–Döring equation with positive excess mass and a macroscopic theory of coarsening is due to Penrose [20]. He observed by formal asymptotics that the macroscopic part of the dynamics converges after a suitable rescaling (cf. Section 2.1) to a classical coarsening model introduced at the same time by Lifshitz and Slyozov [13], and Wagner [23]

$$\begin{aligned} \partial_t \nu_t + \partial_\lambda\big(\lambda^\alpha (u(t) - q\lambda^{-\gamma})\nu_t\big) &= 0 \\ \text{with} \quad u(t) &= \frac{q\int \lambda^{\alpha-\gamma}\,\mathrm{d}\nu_t}{\int \lambda^\alpha\,\mathrm{d}\nu_t}. \end{aligned} \tag{1.10}$$

Hereby, the measure $\nu_t(\mathrm{d}\lambda)$ is the distribution of particles of macroscopic size $\lambda \in \mathbb{R}_+$. Moreover, the parameters α, γ and q satisfy Assumption 1.1 and we will call the nonlocal conservation law (1.10) the LSW equation in the following.

The LSW equation are a gradient flow as formally observed by Niethammer [15, Section 4]. We will make this observation rigorous by using the concept of curves of maximal slope and define a nonnegative functional J characterizing solutions to the LSW equation as its minimizer. The driving energy of the system is given exactly by the first oder expansion of the macroscopic part of the rescaled free energy (1.5) (with $z = z_s$) driving the Becker–Döring equation (cf. Lemma 4.2)

$$E(\nu) := \frac{q}{1-\gamma}\int \lambda^{1-\gamma}\,\nu(\mathrm{d}\lambda). \tag{1.11}$$

An immediate consequence is, that solutions to the LSW equation satisfy an energy-dissipation identity

$$E(\nu_T) - E(\nu_0) + \int_0^T D(\nu_t)\,\mathrm{d}t = 0 \quad \text{with} \quad D(\nu_t) = \int \lambda^\alpha\big(u(t) - q\lambda^{-\gamma}\big)^2\,\mathrm{d}\nu_t, \tag{1.12}$$

where $u(t)$ is given in (1.10).

Passage to the limit. The macroscopic limit is rigorously derived by Niethammer [16]. The main technical tool was to pass to the limit in the energy-dissipation relation associated with the rescaled Becker–Döring equation to obtain the energy-dissipation relation (1.12) of the LSW equation. The one for solutions to the Becker–Döring equation is obtained by integrating the identity (1.6) in time

$$\mathcal{F}(n(T)) - \mathcal{F}(n(0)) + \int_0^T \mathcal{D}(n(t))\,\mathrm{d}t = 0. \tag{1.13}$$

The functional $\mathcal{J}$ from (1.9) contains the identity (1.13), since for solutions of the Becker–Döring equation holds $|n'(z)|^2_{n(t)} = \mathcal{D}(n(t))$.

Let us emphasize, that the proof of [16] is also variational in its nature. We extend this proof and show that the convergence can be lifted to the level of the functionals $\mathcal{J}$ and J That is, a suitable rescaling of the functional $\mathcal{J}^\varepsilon$ converges to the functional J. Hence, the gradient structure of the Becker–Döring equation converges to the one of the LSW equation (cf. Theorem 2.10) and in particular it implies the convergence of solutions (cf. Corollary 2.11). This program will follow the ideas of Sandier and Serfaty [21], and was later generalized by Serfaty [22].

Well preparedness of initial data and quasistationarity. A crucial assumption in the approach of showing convergence via curves of maximal slope is the well preparedness of initial data, which assumes that the rescaled free energy of the Becker–Döring gradient structure converges to the one of the LSW gradient structure

$$\mathcal{F}^\varepsilon(n^\varepsilon(0)) \to E(\nu_0) \qquad \text{as} \qquad \varepsilon \to 0.$$

We will show that on the rescaled time-scale, where the convergence takes place, the Becker–Döring equation reach instantaneously a quasistationary equilibrium, which is dictated only by the monomer concentration. On the other hand, the monomer concentration follows closely a macroscopic quantity similarly defined as u in (1.10). The crucial ingredient in the proof is an energy-dissipation estimate based on a logarithmic Sobolev inequality similarly to the one used in [4] to proof convergence to equilibrium in the noncondensing case $\varrho_0 \leq \varrho_s$.

The quasistationary result shows, that the microscopic part of the rescaled free energy $\mathcal{F}^\varepsilon(n^\varepsilon(t))$ vanishes for almost every $t \geq 0$. This is a first step towards showing, that only the macroscopic part of the rescaled free energy $\mathcal{F}^\varepsilon(n^\varepsilon(0))$ has to convergence towards $E(\nu_0)$ to ensure well prepared initial date. The conjecture is, that the microscopic part is automatically well prepared on the observed rescaled time-scale. This is consistent with the continuous dependence on the initial data of the LSW equation, which is valid under the assumption of convergence of the macroscopic energy for the initial data (see Proposition 2.8).

Outline. The next Section 2 contains in Section 2.1 the gradient flow structure of the Becker–Döring equation and provides the necessary definitions and notations. Moreover, we briefly state the gradient structure of the LSW equation in Section 2.2, which enables us to state the main results in Section 2.3. In Section 3, we prove the gradient flow structure of the LSW equation and prove the continuous dependence on the initial data within this framework. Section 4 contains some a priori estimate for the Becker–Döring system in Section 4.1, therewith we pass then to the limit in the gradient structure in Section 4.2 and finally we prove the quasistationary equilibrium of the small clusters in Section 4.3. We conclude the paper with an Appendix A showing that also more general discrete coagulation and fragmentation models fall into this framework. Moreover, another Appendix B provides an elementary estimate.

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2. Main results

2.1. **The Becker–Döring equation and its gradient structure.**

Discrete gradient flow structures. Based on the interpretation of the Becker–Döring equation as an infinite set of chemical reactions (1.8), we introduce first the gradient structure for reversible chemical reactions. Although the setting in this section is much more general than needed for the introduction of the gradient structure of the Becker–Döring equation, we introduce it on the one hand to make the structure clearer and for the sake of notation. Mielke [14] observed the entropic gradient flow structure for reversible chemical reactions.

Definition 2.1 (Reversible chemical reaction)**.** Let $n \in \mathbb{R}_+^N$ be the densities of $N \in \mathbb{N} \cup \{+\infty\}$ different chemical species (or complexes) X_i reacting according to the mass action law. Each reaction $r = 1, \ldots R$ with $R \in \mathbb{N} \cup \{+\infty\}$ is characterized by the stoichiometric coefficients $x^r, y^r \in \mathbb{N}_0^N$ and forward and backward reaction rates $k_\pm^r > 0$

$$x_1^r X_1 + \cdots + x_N^r X_N \underset{k_-^r}{\overset{k_+^r}{\rightleftharpoons}} y_1^r X_1 + \cdots + y_N^r X_N, \qquad r = 1, \ldots, R. \tag{2.1}$$

The chemical reaction is assumed to be reversible. That is, there exists a state $\omega = (\omega_1, \ldots, \omega_N) \in \mathbb{R}^N$ such that

$$k_+^r \omega^{x^r} = k_-^r \omega^{y^r} =: k^r. \tag{2.2}$$

Here, the notation for multiindices is used: $\omega^{x^r} = \prod_{i=1}^N \omega_i^{x^r_i}$. The evolution equation for the density is given by

$$\dot{n} = -\sum_{r=1}^R k^r \left(\frac{n^{x^r}}{\omega^{x^r}} - \frac{n^{y^r}}{\omega^{y^r}} \right) (x^r - y^r). \tag{2.3}$$

The Becker–Döring clustering equation interpreted as an infinite set of chemical reactions (1.8) fall in this framework by setting $N = R = \infty$ and $x_i^r := \delta_{i,1} + \delta_{i,r}$ and $y_i^r := \delta_{i,r+1}$. The detailed balance condition (2.2) is satisfied in terms of equilibrium distributions $\omega_r(z)$ (1.3), where the choice of the parameter z is still free. Moreover, the more general Smoluchowski coagulation and fragmentation model fit into this framework (cf. Appendix A.1) under the assumption of detailed balance.

The free energy $\mathcal{F}$ is defined as relative entropy with respect to the reversible equilibrium

$$\mathcal{F}(n) := \sum_{l \geq 1} \omega_l \psi\left(\frac{n_l}{\omega_l}\right), \qquad \text{with} \qquad \psi(a) = a \log a - a + 1 \text{ for } a > 0, \tag{2.4}$$

and hence

$$D\mathcal{F}(n) = \left(\log \frac{n_1}{\omega_1}, \ldots, \log \frac{n_i}{\omega_i}, \ldots \right).$$

Note, that $\mathcal{F}$ corresponds to the Lyapunov function (1.5) used for the long-time investigation of the Becker–Döring system.

To define the manifold of states, the stoichiometric subspace and its complement are useful

$$\mathcal{S} := \operatorname{span}\{x^r - y^r : r = 1, \dots R\} \quad \text{and} \quad \mathcal{S}^\perp := \left\{ s \in \mathbb{R}^N : \gamma \cdot s = 0, \ \forall \gamma \in \mathcal{S} \right\}. \tag{2.5}$$

Then, the manifold is given for some fixed $n_0 \in \mathbb{R}_+^N$ by the affine space of densities

$$\mathcal{M}_{n_0} := (n_0 + \mathcal{S}) \cap \mathbb{R}_+^N = \left\{ n \in \mathbb{R}_+^N : n \cdot s = n_0 \cdot s, \forall s \in \mathcal{S}^\perp \right\}$$

The definition formalizes that $\mathcal{S}^\perp$ contains all conversation laws of the reaction and therefore the tangent vectors on $\mathcal{M}_{n_0}$ are given by $\mathbb{R}^{\mathcal{S}}$. Coagulation and fragmentation models of one species, like Becker–Döring, in this terminology are characterized by

$$\mathcal{S}^\perp = \operatorname{span}\{\mathbf{I}\}, \qquad \text{with} \qquad \mathbf{I} := (1, 2, 3, 4, \dots).$$

Hence, the manifold has only one conserved quantity, which is the density $\varrho_0 > 0$ of the total number of particles

$$\mathcal{M} := \mathcal{M}_{\varrho_0} = \left\{ n \in \mathbb{R}_+^{\mathbb{N}} : n \cdot \mathbf{I} = \sum_{l=1}^\infty l n_l = \varrho_0 \right\}.$$

The derivative of the energy $D\mathcal{F}$ is a force and has to be interpreted as covector. The underlying metric can be specified by mapping covectors to (tangent-)vectors. This is done via the Onsager matrix to be defined as the symmetric semi-positive definite matrix

$$\mathcal{K}(n) := \sum_r k^r \Lambda\left(\frac{n^{x^r}}{\omega^{x^r}}, \frac{n^{y^r}}{\omega^{y^r}} \right) (x^r - y^r) \otimes (x^r - y^r), \tag{2.6}$$

where $\Lambda(\cdot, \cdot)$ is the logarithmic mean defined for $a, b > 0$ by

$$\Lambda(a, b) = \int_0^1 a^s b^{1-s} \, \mathrm{d}s = \begin{cases} \frac{a-b}{\log a - \log b} & , a \neq b \\ a & , a = b. \end{cases}$$

Hence, recalling that the space of vectors was given by $\mathbb{R}^{\mathcal{S}}$, we define the covectors with the help of the Onsager operator by

$$\mathcal{T}_n^* \mathcal{M}_{n_0} := \left\{ \phi \in \mathbb{R}^N : \exists s \in \mathbb{R}^{\mathcal{S}} \text{ such that } s = -\mathcal{K}(n)\phi \right\},$$

where the identification is well-defined since $\mathcal{K}$ is by definition strictly positive definite on $\mathbb{R}^{\mathcal{S}}$, whenever n is strictly positive in all of its components. Note, although the tangent space is state independent, this is not the case for the cotangent space.

With this preliminary definitions a reversible chemical reaction as given in Definition 2.1 is formally the gradient flow of the free energy $\mathcal{F}$ (2.4) with respect to the metric structure induced by the Onsager operator (2.6) and it holds the formal identity

$$\dot{n} = -\mathcal{K}(n) D\mathcal{F}(n). \tag{2.7}$$

The property from which immediately follows that (2.7) is the same as (2.3) is

$$(x^r - y^r) \cdot D\mathcal{F}(n) = \sum_{i=1}^n x_i^r \log \frac{n_i}{\omega_i} - y_i^r \log \frac{n_i}{\omega_i} = \log \frac{n^{x^r}}{\omega^{x^r}} - \log \frac{n^{y^r}}{\omega^{y^r}},$$

which is nothing else than the nominator of the logarithmic mean $\Lambda\big(\frac{n^{x^r}}{\omega^{x^r}}, \frac{n^{y^r}}{\omega^{y^r}}\big)$ and resembles a discrete chain rule. The gradient flow decreases its energy along its evolution in terms of the dissipation, i.e.

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{F}(n) &= D\mathcal{F}(n)\cdot\dot{n} = -D\mathcal{F}(n)\cdot\mathcal{K}(n)D\mathcal{F}(n)\\ &= -\sum_r k^r\left(\frac{n^{x^r}}{\omega^{x^r}}-\frac{n^{y^r}}{\omega^{y^r}}\right)\left(\log\frac{n^{x^r}}{\omega^{x^r}}-\log\frac{n^{y^r}}{\omega^{y^r}}\right) =: -\mathcal{D}(n)\end{aligned}$$

We see that the Becker–Döring system fits into this framework. However, there is freedom in the choice of the free energy and under certain physical assumption, there are other possible choices and in the Appendix A.2, we show that also a modified Becker–Döring system introduced by [8] can be brought into the form of a gradient flow.

Curves of finite action. A crucial ingredient to study the underly metric structure of a gradient flow, is the continuity equation and curves of finite action.

Definition 2.2 (Continuity equation)**.** A pair $[0,T]\ni t\mapsto(n(t),\phi(t))\in\mathcal{M}_{n_0}\times\mathcal{T}^*_{n(t)}\mathcal{M}_{n_0}$ is a solution to the continuity equation, denoted by $(n,\phi)\in\mathcal{CE}_T$, if it satisfies

(i) $n(\cdot):[0,T]\to\mathcal{M}_{n_0}$ is absolute continuous
(ii) $n(t)\in\mathcal{M}_{n_0}(\mathcal{X})$ for all $t\in[0,T]$
(iii) The pair (n,ϕ) satisfies the continuity equation for $t\in(0,T)$ in the weak form, that is for all $\psi\in C^1_c((0,T),\mathbb{R})$ and all $l\in\{1,\dots,N\}$ holds

$$\int_0^T\left(\dot\psi(t)\;n_l(t)-\psi(t)\;\big(\mathcal{K}(n(t))\phi(t)\big)_l\right)\mathrm{d}t = 0. \tag{2.8}$$

Definition 2.3 (Curves of finite action)**.** The action $\mathcal{A}$ of a pair $(n,\phi)\in\mathcal{M}_{n_0}\times\mathcal{T}^*_n\mathcal{M}_{n_0}$ is defined by

$$\mathcal{A}(n,\phi) := \phi(t)\cdot\mathcal{K}(n(t)\phi(t) = \sum_{r=1}^R k^r\hat{n}^\omega_r\left|\nabla_r\phi(t)\right|^2, \tag{2.9}$$

where $\nabla_r\phi := (y^r-x^r)\cdot\phi = \phi_{r+1}-\phi_r-\phi_1$. The *dissipation* for $n\in\mathcal{M}_{n_0}$ is defined by

$$\mathcal{D}(n) := \mathcal{A}(n,-D\mathcal{F}(n)).$$

A curve $(n,\phi)\in\mathcal{CE}_T$ is called a *curve of finite action*, if

$$\sup_{t\in[0,T]}\mathcal{F}(n(t))<\infty,\qquad\int_0^T\mathcal{A}(n(t),\phi(t))\,\mathrm{d}t<\infty,\quad\text{and}\qquad\int_0^T\mathcal{D}(n(t))\,\mathrm{d}t<\infty$$

Remark 2.4. It is possible to define a discrete Wasserstein metric $\mathcal{W}$ for $n^0,n^1\in\mathcal{M}_{n_0}(\mathcal{X})$ by the Benamou–Brenier formula

$$\mathcal{W}(n^0,n^1)^2 := \inf\left\{\int_0^1\mathcal{A}(n(t),\phi(t))\,\mathrm{d}t : (n,\phi)\in\mathcal{CE}(n^0,n^1)\right\},$$

where $(n,\phi)\in\mathcal{CE}(n^0,n^1)$ are solutions to the continuity with $T=1$ and initial and terminal state $n(0)=n^0$ and $n(1)=n^1$, respectively. In settings similar to the present, this was done in [9] and [10]. The metric would allow to define absolute continuous curves and the metric velocity in the metric space $(\mathcal{M}_{n_0},\mathcal{W})$. However, for the pure convergence statement, it is enough to work in the class of curves of finite action, which by definition of the above metric are also absolute continuous in this setting.

Curves of finite action can give a variational formulation to solutions of chemical reactions and in particular to the Becker–Döring equation. In comparison to the direct gradient flow equation (2.7), this avoids regularity questions arising from the application of the chain rule. The concept first appeared in [6] and was rigorously investigated in [1].

Proposition 2.5 (Curves of maximal slope)**.** *For any curve $(n, \phi) \in \mathcal{CE}_T$ of finite action holds*

$$\mathcal{J}(n) := \mathcal{F}(n_T) - \mathcal{F}(n_0) + \frac{1}{2}\int_0^T \mathcal{D}(n_t)\ \mathrm{d}t + \frac{1}{2}\int_0^T \mathcal{A}(n_t, \phi_t)\ \mathrm{d}t \geq 0.$$

Moreover, equality is attained if and only if n is a solution of (2.3)*.*

Sketch of the proof. We provide the crucial observation of the proof, which follows formally by evaluating

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{F}(n(t)) &= D\mathcal{F}(n(t)) \cdot \dot{n}(t) \overset{(2.8)}{=} D\mathcal{F}(n(t)) \cdot \mathcal{K}(n(t))\phi(t) \\ &\geq -\frac{1}{2} D\mathcal{F}(n(t)) \cdot \mathcal{K}(n(t)) D\mathcal{F}(n(t)) - \frac{1}{2}\phi(t) \cdot \mathcal{K}(n(t))\phi(t),\end{aligned}$$

where we used that $\mathcal{K}$ is positive semidefinite and the Cauchy–Schwarz inequality. The equality case is read off from the equality case in Cauchy–Schwarz. For a rigorous treatment in a similar situation, we refer to [10, Section 2.5]. □

Heuristics and scaling. From now, we consider the Becker–Döring system with initial total mass $\varrho_0 > \varrho_s$ and rates satisfying Assumption 1.1. Moreover, the reference state for the free energy is given by $\omega = \omega(z_s)$ as defined in (1.3).

To obtain the necessary rescaling, we will use the result by [2] that the free energy $\mathcal{F}$ (2.4) is decreasing to 0 as $t \to \infty$. Then, we expect that if the energy is small the scale of the large cluster should be given by some power of the energy. It will be more convenient to answer to consider this observation the other way around like done by [16]. We fix a scale ε^{-1} with $\varepsilon > 0$ for the large cluster and ask for the first order expansion of the energy in ε. Therefore, for all l larger some cut-off l_0, we introduce $\lambda = \varepsilon l$ and treat λ as continuous variable.

We rescale the cluster density n_l by ε^2 and define the empirical measure

$$\nu^\varepsilon(d\lambda) := \varepsilon \sum_{l \geq l_0} \delta_{\varepsilon l}(\lambda) \frac{n_l}{\varepsilon^2} = \frac{1}{\varepsilon} \sum_{l \geq l_0} \delta_{\varepsilon l}(\lambda) n_l \qquad \text{in } C_c^0(\mathbb{R})^*. \tag{2.10}$$

That is for each $\zeta \in C_c^0(\mathbb{R})$ holds

$$\int_0^\infty \zeta(\lambda)\, \nu^\varepsilon(\mathrm{d}\lambda) = \frac{1}{\varepsilon} \sum_{l \geq l_0} \zeta(\varepsilon l) n_l.$$

This scaling preserves the mass in the large cluster, which follows by approximating $\zeta(\lambda) = \lambda$ with cut-off functions.

We also want to make a statement on the behavior of the small clusters and introduce the according truncated distribution,

$$m_l^\varepsilon = \begin{cases} n_l & , 1 \leq l < l_0 \\ 0 & , l \geq l_0 \end{cases}. \tag{2.11}$$

Now, with the rates given in Assumption 1.1, we expect the leading order contribution of the free energy to be given by the free energy of the large clusters. This part of the free energy (2.4) can be expanded (cf. Lemma 4.2) as follows

$$\begin{aligned}\mathcal{F}(n) \geq \mathcal{F}_{\mathrm{mac}}(n) &:= \sum_{l\geq l_0} \omega_l \psi\left(\frac{n_l}{\omega_l}\right)\\ &= \left(\frac{q}{z_s(1-\gamma)} \sum_{l\geq l_0} l^{1-\gamma} n_l\right)\left(1+O(l_0^{-\sigma})+O(l_0^{\gamma} w_{l_0})\right)\\ &= \frac{\varepsilon^{\gamma} q}{z_s(1-\gamma)} \int \lambda^{1-\gamma}\,\mathrm{d}\nu^{\varepsilon}\left(1+O(l_0^{-\sigma})+O(l_0^{\gamma} w_{l_0})\right), \end{aligned} \tag{2.12}$$

for some $\sigma > 0$. Hence, we define the rescaled free energy as

$$\mathcal{F}^{\varepsilon}(n) = \frac{1}{\varepsilon^{\gamma}} \mathcal{F}(n)$$

and split it into microscopic and macroscopic free energy as

$$\mathcal{F}^{\varepsilon}_{\mathrm{mic}}(m^{\varepsilon}) := \frac{1}{\varepsilon^{\gamma}} \sum_{l=1}^{l_0-1} \omega_l \psi\left(\frac{n_l}{\omega_l}\right) \qquad \text{and} \qquad \mathcal{F}^{\varepsilon}_{\mathrm{mac}}(\nu^{\varepsilon}) := \frac{1}{\varepsilon^{\gamma}} \sum_{l\geq l_0} \omega_l \psi\left(\frac{n_l}{\omega_l}\right).$$

So the rescaled free energy is of order 1, whenever the free energy is of order ε^{γ}. Since, we already know that the total free energy decreases to zero as $t \to \infty$, we could introduce a time t_{ε} such that $\mathcal{F}(n(t_{\varepsilon})) = O(\varepsilon^{\gamma})$ and treat $n(t_{\varepsilon})$ as initial value. This implies that for ε small, all possible existing metastable states are already broken down (cf. [19]).

The expansion (2.12) also shows, that the cut-off l_0 has to satisfy two conditions (cf. (4.2) and (4.4))

$$\lim_{\varepsilon\to 0} l_0^{\gamma} w_{l_0} = 0 \qquad \text{and} \qquad \lim_{\varepsilon\to 0} \max\{l_0^{\gamma}, l_0^{\alpha}\} \sqrt{\mathcal{F}(n^{\varepsilon}(0))} = 0.$$

The first ensures, that it is large enough to capture the tail of the equilibrium distribution and the second is a compatibility condition with the initial distribution. By taking into account the asymptotic of $\{Q_l\}_{l\geq 1}$ (cf. Lemma 4.1) and recalling $w_l = z_s^l Q_l$, the cut-off l_0 can be chosen as

$$l_0 := \lfloor \varepsilon^{-x} \rfloor \qquad \text{for some} \qquad x \in \left(0, \tfrac{1}{2}\right). \tag{2.13}$$

We consider only states n such that free energy is of order ε^{γ}, that is we consider the restricted state space

$$\mathcal{M}^{\varepsilon} := \left\{ n \in \mathbb{R}^{\infty}_{+} : \sum_{l\geq 1} l n_l = \varrho_0 \text{ and } \mathcal{F}(n) \leq \varepsilon^{\gamma} \right\}.$$

On this state space, we have two projections

$$\begin{aligned} \Pi^{\varepsilon}_{\mathrm{mac}} : \mathcal{M}^{\varepsilon} &\to \mathcal{M}^{\varepsilon}_{\mathrm{mac}} & \qquad \Pi^{\varepsilon}_{\mathrm{mic}} : \mathcal{M}^{\varepsilon} &\to \mathcal{M}^{\varepsilon}_{\mathrm{mic}}\\ n &\mapsto \nu^{\varepsilon} & n &\mapsto m^{\varepsilon}, \end{aligned}$$

where ν^{ε} and m^{ε} are defined in (2.10) and (2.11), respectively and $l_0 = \lfloor \varepsilon^{-x} \rfloor$ as in (2.13). The according state spaces $\mathcal{M}^{\varepsilon}_{\mathrm{mac}}$ and $\mathcal{M}^{\varepsilon}_{\mathrm{mic}}$ are just the image of $\Pi^{\varepsilon}_{\mathrm{mac}}$ and $\Pi^{\varepsilon}_{\mathrm{mic}}$, respectively. Likewise, the differential of the free energy for states $n^{\varepsilon} \in \mathcal{M}^{\varepsilon}$ will be of order ε^{γ} and hence covectors will be also on scale ε^{γ}, that is we define a rescaled vector field w^{ε} by

$$\nabla_l \phi = (y_l - x_l) \cdot \phi = \phi_{l+1} - \phi_l - \phi_1 =: \varepsilon^{\gamma} w^{\varepsilon}(\varepsilon l). \tag{2.14}$$

The rescaling of tangent vectors is then determined by the rescaling necessary for obtaining the macroscopic Onsager operator $\mathcal{K}$. This follows heuristically by expanding the Onsager operator

$$(\mathcal{K}(n)\phi)_l = -\varepsilon^{1-\alpha}\partial^\varepsilon_\lambda(z_s\lambda^\alpha\varepsilon^\gamma w^\varepsilon)(1+O(\varepsilon^{\alpha\gamma})) \quad \text{with} \quad \partial^\varepsilon_\lambda f(\lambda) := \frac{f(\lambda+\varepsilon)-f(\lambda)}{\varepsilon}.$$

Formally, ε turns the discrete derivative in a continuous one and ε^α comes from the additional weight λ^α. Hence, we define the rescaled Onsager operator by

$$(\mathcal{K}^\varepsilon(n)w^\varepsilon)(\varepsilon l) := \frac{1}{\varepsilon^{1-\alpha+\gamma}}(\mathcal{K}(n)\phi)(l),$$

where w^ε and ϕ are given by the relation (2.14). This rescaling translates to the action $\mathcal{A}(n,\phi) = \phi\cdot\mathcal{K}(n)\phi$ and we define the rescaled action by

$$\mathcal{A}^\varepsilon(n,w^\varepsilon) := \frac{1}{\varepsilon^{1-\alpha+2\gamma}}\sum_{l\geq 1} k^l \hat{n}^\omega_l |\nabla_l\phi|^2. \tag{2.15}$$

Since, the dissipation is given as $\mathcal{D}(n) := \mathcal{A}(n, -D\mathcal{F}(n))$, the rescaling is the same and we define

$$\mathcal{D}^\varepsilon(n) = \frac{1}{\varepsilon^{1-\alpha+2\gamma}}\sum_{l\geq 1} k^l \hat{n}^\omega_l |\nabla_l D\mathcal{F}(n)|^2$$

Hence the total rescaling between cotangent and tangent vectors is $\varepsilon^{1-\alpha+\gamma}$, which fixes the time scale for the macroscopic process.

Now, we introduce rescaled curves of finite action in analog to Definitions 2.2 and 2.3. By abuse of notation the new time-scale $t/\varepsilon^{1-\alpha+\gamma}$ is still denoted by t.

Definition 2.6 (Rescaled curves of finite action)**.** A weak solution $[0,T]\ni t\mapsto (n^\varepsilon(t), w^\varepsilon(t))$ to the rescaled continuity equation

$$\int_0^T \Big(\dot{\psi}(t)n^\varepsilon_l(t) - \psi(t)(\mathcal{K}^\varepsilon(n^\varepsilon(t))w^\varepsilon(t))_l\Big)\,\mathrm{d}t = 0, \qquad \text{for all } \psi\in C^1_c((0,T);\mathbb{R})$$

denoted by $(n^\varepsilon, w^\varepsilon)\in\mathcal{CE}^\varepsilon_T$ is called a rescaled curve of finite action if

$$\sup_{t\in[0,T]}\mathcal{F}^\varepsilon(\nu^\varepsilon_t) < \infty, \quad \int_0^T \mathcal{A}^\varepsilon(n^\varepsilon(t), w^\varepsilon(t))\;\mathrm{d}t < \infty \quad \text{and} \quad \int_0^T \mathcal{D}^\varepsilon(n^\varepsilon(t))\;\mathrm{d}t < \infty.$$

Moreover, for such a curve we define the rescaled functional characterizing curves of maximal slope by

$$\mathcal{J}^\varepsilon(n^\varepsilon) := \mathcal{F}^\varepsilon(n^\varepsilon(T)) - \mathcal{F}^\varepsilon(n^\varepsilon(0)) + \frac{1}{2}\int_0^T \mathcal{D}^\varepsilon(n^\varepsilon(t))\,\mathrm{d}t + \frac{1}{2}\int_0^T \mathcal{A}^\varepsilon(n^\varepsilon(t), w^\varepsilon(t))\,\mathrm{d}t \geq 0. \tag{2.16}$$

In particular solutions such that $\mathcal{J}^\varepsilon(n^\varepsilon) = 0$ satisfy the rescaled Becker–Döring equation

$$\dot{n}^\varepsilon(t) = \mathcal{K}^\varepsilon(n^\varepsilon(t))D\mathcal{F}(n^\varepsilon(t)) = \varepsilon^{1-\alpha+\gamma}\mathcal{K}(n^\varepsilon(t))\mathcal{D}\mathcal{F}(n^\varepsilon(t)). \tag{2.17}$$

The last ingredient is an additional tightness assumption on the initial data, which ensures certain tightness properties.

Assumption 2.7. *A family $\{\nu^\varepsilon\}_{\varepsilon>0}$ of nonnegative Radon measure on $\mathbb{R}_+$ is tight, provided that*

$$\int_R^\infty \lambda\,\nu^\varepsilon_0(\mathrm{d}\lambda)\to 0 \qquad \textit{as } R\to\infty \qquad \textit{uniformly in } \varepsilon.$$

2.2. **The macroscopic limiting model: LSW equation.** The parameters $\alpha \in [0,1), \gamma \in (0,1)$ and $\varrho_0 > \varrho_s > 0$ occurring in the below expressions are fixed by Assumption 1.1 and the initial data of Becker–Döring equation. Hence, we do not explicitly point them out in the following.

The LSW equation model the cluster growth and are the solution to the following nonlocal conservation law

$$\partial_t \nu_t + \partial_\lambda\big(\lambda^\alpha\big(u(\nu_t) - q\lambda^{-\gamma}\big)\nu_t\big) = 0 \qquad \text{in } C_c^\infty([0,T]\times\mathbb{R}_+)^* \tag{2.18}$$

$$u(\nu_t) = \frac{q\int \lambda^{\alpha-\gamma}\,\nu_t(\mathrm{d}\lambda)}{\int \lambda^\alpha\,\nu_t(\mathrm{d}\lambda)}. \tag{2.19}$$

Formally, the total mass is conserved and it holds for $t > 0$

$$\int \lambda\,\nu_t(\mathrm{d}\lambda) = \int \lambda\,\nu_0(\mathrm{d}\lambda) =: \varrho_0 - \varrho_s =: \bar{\varrho}.$$

Therefore, the densities on $\mathbb{R}_+ := (0,\infty)$ are confined to

$$M = \left\{ \nu \in C_c^0(\mathbb{R}_+)^* \;\middle|\; \int \lambda\,\nu(\mathrm{d}\lambda) = \bar{\varrho} \right\}.$$

Let us introduce a formal Riemannian structure and define a tangent space on M by

$$T_\nu M := \left\{ s : \mathbb{R}_+ \to \mathbb{R} \;\middle|\; \int \lambda s\,\mathrm{d}\lambda = 0 \right\}$$

A possible identification of cotangent vectors is via the usual Wasserstein gradient, given in terms of the Onsager operator $\tilde{K}(\nu) : \tilde{T}_\nu^* M \to T_\nu M$ by $\tilde{K}(\nu)v := -\partial_\lambda(v\nu)$. Hence, we define

$$\tilde{T}_\nu^* M := \left\{ v : \mathbb{R}_+ \to \mathbb{R} \;\middle|\; \exists s \in T_\nu M : \tilde{K}(\nu)v = s \right\}.$$

Note, that the condition $\int \lambda s\,\nu(\mathrm{d}\lambda) = 0$ is necessary for the existence of a solution to $K(\nu)v = s$. Therewith, the metric tensor is defined by

$$\tilde{g}_\nu(s_1,s_2) := \int \frac{v_1 v_2}{\lambda^\alpha}\,\nu(\mathrm{d}\lambda) \quad \text{with} \quad s_i = -\tilde{K}(\nu)v_i \text{ in } C_c^\infty(\mathbb{R}_+)^* \quad \text{for } i = 1,2. \tag{2.20}$$

In our case, it will be convenient to work with the slightly changed cotangent space, which will be a more direct limit of the discrete structure. Therefore, the identification of tangent and cotangent vectors is obtained via the operator $K(\nu) : T_\nu^* M \to T_\nu M$ defined by $K(\nu)w := -\partial_\lambda(\lambda^\alpha w\nu)$ for $w \in T_\nu^* M$. Therewith, the cotangent space is given by

$$\begin{aligned} T_\nu^* M &:= \{ w : \mathbb{R}_+ \to \mathbb{R} \mid \exists s \in T_\nu M : K(\nu)v = s \} \\ &\subseteq \left\{ w : \mathbb{R}_+ \to \mathbb{R} \;\middle|\; \int \lambda^\alpha w\,\nu(\mathrm{d}\lambda) = 0 \right\}, \end{aligned}$$

where the inclusion property follows by integration by parts of the identity $0 = \int \lambda s\,\nu(\mathrm{d}\lambda)$. Furthermore, the metric is now given by

$$g_\nu(s_1,s_2) := \int \lambda^\alpha w_1 w_2\,\nu(\mathrm{d}\lambda) \quad \text{with} \quad s_i = -K(\nu)w_i \text{ in } C_c^\infty(\mathbb{R}_+)^* \quad \text{for } i = 1,2.$$

The energy is modeled as a surface energy and given by (1.11).

Therewith, let us formally derive the gradient structure for the LSW equation (cf. [15, Section 4]), that is we assume all differentials and quantities to be smooth enough. The differential of the energy (1.11) is given for some $s \in T_\nu M$ by using the identification $s = -K(\nu)w$ with $w \in T^*_\nu M$

$$DE(\nu) \cdot s = \frac{q}{1-\gamma} \int \lambda^{1-\gamma} s \, \mathrm{d}\lambda = - \int \Big(u\lambda - \frac{q}{1-\gamma} \lambda^{1-\gamma} \Big) s \, \nu(\mathrm{d}\lambda)$$
$$= - \int \lambda^\alpha (u - q\lambda^{-\gamma}) w \, \nu(\mathrm{d}\lambda),$$

where $u \in \mathbb{R}$ can be chosen arbitrary thanks to $\int \lambda^\alpha w \nu(\mathrm{d}\lambda)$. Then, the gradient flow in weak form satisfies for all $\tilde{s} \in T_\nu M$

$$\int \lambda^\alpha w \tilde{w} \, \nu(\mathrm{d}\lambda) = g_{\alpha,\nu}(\partial_t \nu, \tilde{s}) = -DE(\nu) \cdot \tilde{s} = \int \lambda^\alpha (u - q\lambda^{-\gamma}) \tilde{w} \, \nu(\mathrm{d}\lambda),$$

where $\partial_t \nu = -\partial_\lambda(\lambda^\alpha w \nu)$ and $\tilde{s} = -\partial_\lambda(\lambda^\alpha \tilde{w} \nu)$ in distribution. Hence, we obtain the identification

$$w = u - q\lambda^{-\gamma},$$

where $u = u(\nu)$ is a Lagrangian multiplier chosen such that $w \in T^*_\nu M$, that is it satisfies the constraint $\int \lambda^\alpha w \, \nu(\mathrm{d}\lambda) = 0$ and is formally given by (2.19) So the gradient flow of the energy E with respect to the metric induced by K is given by

$$\partial_t \nu_t = -K(\nu) DE(\nu) = -\partial_\lambda \big(\lambda^\alpha \big(u(\nu_t) - q\lambda^{-\gamma} \big) \nu_t \big),$$

where $u(\nu_t)$ given by (2.19). Differentiating the energy along a solution yields the dissipation, which takes the form

$$-\frac{\mathrm{d}}{\mathrm{d}t} E(\nu) = - \int \lambda^\alpha \big(u(\nu_t) - q\lambda^{-\gamma} \big)^2 \nu_t(\mathrm{d}\lambda) =: -D(\nu). \tag{2.21}$$

To make the above observation rigorous, we will use the de Giorgi formalism of curves of maximal slope. Up to technical details, which will be dealt with in Section 3, we can define an action functional as follows: For a pair (ν, w) solving the continuity equation $\partial_t \nu_t + \partial_\lambda(\lambda^\alpha w_t \nu_t) = 0$ in distributions, denoted by $(\nu, w) \in \mathrm{CE}_T$, the action is defined by

$$\int_0^T A(\nu_t, w_t) \, \mathrm{d}t \qquad \text{with} \qquad A(\nu_t, w_t) = \int \lambda^\alpha |w_t|^2 \, \mathrm{d}\nu_t.$$

Then, by the identification of tangent and co-tangent vectors via $s = -\partial_\lambda(\lambda^\alpha w \nu)$, we obtain that the dissipation is given by

$$D(\nu_t) = A(\nu_t, u(t) - DE(\nu_t)) = \int \lambda^\alpha \big| u(\nu_t) - q\lambda^{-\gamma} \big|^2 \, \mathrm{d}\nu_t,$$

where $u(\nu_t) \in L^2((0,T))$ given by (2.19) ensures that $u(\nu_t) - q\lambda^{-\gamma} \in T^*_\nu M$ is a valid cotangent vector satisfying $\int \lambda^\alpha (u(\nu_t) - q\lambda^{-\gamma}) \, \mathrm{d}\lambda = 0$.

Therewith, the functional $J(\nu)$, which completely characterizes solutions to (2.18) (cf. Proposition 3.7) is defined by

$$J(\nu) := E(\nu_T) - E(\nu_0) + \frac{1}{2} \int D(\nu_t) \, \mathrm{d}t + \frac{1}{2} \int A(\nu_t, w_t) \, \mathrm{d}t \geq 0, \tag{2.22}$$

with $J(\nu) = 0$ if and only if ν_t is a weak solution to the LSW equation (2.18).

The above characterization of solution to the LSW equation together with a compactness statement for curves of finite action (cf. Proposition 3.6) allows to proof continuous dependence on the initial data within the gradient flow framework.

Proposition 2.8 (Continuous dependency on the initial data). *Let $\{\nu_0^\varepsilon\}_{\varepsilon>0}$ be a sequence of initial data satisfying the tightness Assumption 2.7 and*

$$\lim_{\varepsilon \to 0} E(\nu_0^\varepsilon) = E(\nu_0) < \infty. \tag{2.23}$$

Then there exists a solution $\nu \in C_c^\infty([0,T] \times \mathbb{R}_+)^*$ to the LSW equation such that $\nu_t^\varepsilon \overset{*}{\rightharpoonup} \nu_t$ in $C_c^0(\mathbb{R}_+)$ for all $t \in [0,T]$.

Remark 2.9. The above result is consistent with the existing literature: In [18, Theorem 2.2], the continuous dependency on the initial data was shown under Assumption 2.7 with respect to weak* convergence for continuous test functions compactly supported on $[0,\infty)$ including 0, i.e. Borel measures on $[0,\infty)$. Then, it is easy to see that weak* convergence with respect to this class implies convergence of the macroscopic energy (2.33).

2.3. **Convergence of the gradient structures.** The functionals $\mathcal{J}^\varepsilon$ (2.16) and J (2.22) are used to characterize solutions of the Becker–Döring and LSW equations in a variational way, respectively. The main idea to show convergence of the Becker–Döring equation to the LSW equation, which goes back to [21] (cf. [22]), is to prove $\liminf_{\varepsilon\to 0}\mathcal{J}^\varepsilon(n^\varepsilon)\geq J(\nu)$ for curves of finite action n^ε converging to ν. The lower semi-continuity estimate can be established by showing individual semi-continuity estimates for the energy, action and dissipation. This is the content of Theorem 2.10.

Theorem 2.10 (Convergence of curves of finite action)**.** *Suppose that* $\alpha\geq 1-3\gamma$*. For* $T>0$ *let* $(n^\varepsilon,w^\varepsilon)\in\mathcal{CE}^\varepsilon_T$ *be a curve of finite action and* $\nu_0^\varepsilon:=\Pi^\varepsilon_{\mathrm{mac}}n^\varepsilon(0)$ *satisfy the Assumption 2.7, then there exists a limiting curve* $t\mapsto(\nu_t,w_t)\in\mathrm{CE}_T$ *such that*

$$\nu_t^\varepsilon:=\Pi^\varepsilon_{\mathrm{mac}}n^\varepsilon(t)\overset{*}{\rightharpoonup}\nu_t \qquad \text{in } C^0_c(\mathbb{R}_+)^* \qquad \text{for all } t\in[0,T] \tag{2.24}$$

and

$$w_t^\varepsilon(\lambda)\nu_t^\varepsilon(\mathrm{d}\lambda)\,\mathrm{d}t\overset{*}{\rightharpoonup}w_t(\lambda)\,\mathrm{d}\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t \qquad \text{in} \quad C^0_c([0,T]\times\mathbb{R}_+)^*. \tag{2.25}$$

There exists $u\in L^2((0,T))$ *such that*

$$h^\varepsilon(t):=\frac{n_1(t)-z_s}{\varepsilon^\gamma}\rightharpoonup u(t), \qquad \text{weakly in } L^2((0,T)), \tag{2.26}$$

and $u(t)$ *satisfies the identity*

$$u(t)=\frac{q\int\lambda^{\alpha-\gamma}\,\nu_t(\mathrm{d}\lambda)}{\int\lambda^\alpha\,\nu_t(\mathrm{d}\lambda)}.$$

Moreover, the energy, the action and the dissipation satisfy the following lim inf *estimates*

$$\forall t\in[0,T]: \qquad \lim_{\varepsilon\to 0}\mathcal{F}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon)\geq\frac{1}{z_s}E(\nu_t), \tag{2.27}$$

$$\liminf_{\varepsilon\to 0}\int_0^T\mathcal{A}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon,w_t^\varepsilon)\,\mathrm{d}t\geq\frac{1}{z_s}\int_0^T A(\nu_t,w_t)\,\mathrm{d}t, \tag{2.28}$$

$$\liminf_{\varepsilon\to 0}\int_0^T\mathcal{D}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon)\,\mathrm{d}t\geq\frac{1}{z_s}\int_0^T D(\nu_t)\,\mathrm{d}t. \tag{2.29}$$

The classical conclusion from the above theorem is the convergence of curves of maximal slope under the assumption of well-prepared initial data to deal with the term $-\mathcal{F}^\varepsilon(n^\varepsilon(0))$ inside of $\mathcal{J}^\varepsilon(n^\varepsilon)$. The following Corollary is an immediate consequence of Theorem 2.10 by the arguments of [22, Theorem 2].

Corollary 2.11 (Convergence of curves of maximal slope)**.** *Suppose* $\alpha\geq 1-3\gamma$ *and let* $(n^\varepsilon,w^\varepsilon)\in\mathcal{CE}^\varepsilon_T$ *be a curve of finite action. Moreover assume* $\nu_0^\varepsilon:=\Pi^\varepsilon_{\mathrm{mac}}n^\varepsilon(0)$ *satisfy the Assumption 2.7 and* $n^\varepsilon(0)$ *is well-prepared in the sense that*

$$\lim_{\varepsilon\to 0}\mathcal{F}^\varepsilon(n^\varepsilon(0))=E(\nu_0).$$

Then, there exists a limiting $(\nu,w)\in\mathrm{CE}_T$ *satisfying* (2.24) *and* (2.25) *such that*

$$\liminf_{\varepsilon\to 0}\mathcal{J}^\varepsilon(n^\varepsilon)\geq J(\nu)\geq 0.$$

Especially, if $\mathcal{J}^\varepsilon(n^\varepsilon) = 0$ then $J(\nu) = 0$ and it holds

$$\begin{aligned} \lim_{\varepsilon\to 0} \mathcal{F}^\varepsilon(n^\varepsilon(t)) &= \frac{1}{z_s} E(\nu_t) && \text{for all } t \in [0,T], \\ \mathcal{A}^\varepsilon(n^\varepsilon, w^\varepsilon) &\to \frac{1}{z_s} A(\nu, w) && \text{for a.e. } t \in [0,T], \\ \mathcal{D}^\varepsilon(n^\varepsilon) &\to \frac{1}{z_s} D(\nu) && \text{for a.e. } t \in [0,T]. \end{aligned}$$

The statement (2.26) connects the microscopic monomer concentration with a ratio of moments of the macroscopic cluster distribution. It is possible to show this identity already on the level of rescaled Becker–Döring equation alone. That is, the monomer concentration follows closely a moment ratio of the distribution of the large clusters.

Proposition 2.12. *For any curve $(n^\varepsilon, \phi^\varepsilon) \in \mathcal{CE}_T^\varepsilon$ such that $\mathcal{J}^\varepsilon(n^\varepsilon) < \infty$ uniformly in ε and ν_0^ε satisfying Assumption 4.5 the rescaled monomer excess concentration h^ε as defined in (2.26) satisfies*

$$\int_0^T \left(h^\varepsilon(t) - u^\varepsilon(t)\right)^2 dt \le C \int_0^T \mathcal{D}^\varepsilon_{\mathrm{mac}}(n(t))\, dt, \tag{2.30}$$

with

$$u^\varepsilon(t) := \frac{\sum_{l\ge l_0} (b_{l+1} n_{l+1}(t) - a_l n_l)}{\varepsilon^\gamma \sum_{l\ge l_0} a_l n_l}.$$

The above results together with a refined energy-dissipation estimate based on a logarithmic Sobolev inequality allows to establish detailed information on the distribution of the small clusters for curves of rescaled finite action and in particular for every solution of the time-rescaled Becker–Döring equation (2.17). The result makes part of the formal asymptotic contained in [16, Section 3] rigorous.

Theorem 2.13 (Quasistationary distribution)**.** *For any curve $(n^\varepsilon, \phi^\varepsilon) \in \mathcal{CE}_T^\varepsilon$ such that $\mathcal{J}^\varepsilon(n^\varepsilon) < \infty$ uniformly in ε and ν_0^ε satisfying Assumption 4.5 the small cluster follow a quasistationary distribution dictated by n_1: For $l_0 = \lfloor \varepsilon^{-x} \rfloor$ with x satisfying (2.13) holds*

$$\int_0^T \mathcal{H}_{\mathrm{mic}}\bigl(n^\varepsilon(t) \mid \omega(n_1^\varepsilon(t))\bigr)\,\mathrm{d}t \le C \varepsilon^{\gamma+(1-x)(1-\alpha+\gamma)} \int_0^T \mathcal{D}^\varepsilon_{\mathrm{mic}}(n_t^\varepsilon)\,\mathrm{d}t, \tag{2.31}$$

where $\omega_l(z) = z^l Q_l$ as defined in (1.3) and $\mathcal{H}_{\mathrm{mic}}$ is the relative entropy defined by

$$\mathcal{H}_{\mathrm{mic}}(n \mid \omega(z)) := \sum_{l=1}^{l_0-1} \omega_l(z) \psi\left(\frac{n_l}{\omega_l(z)}\right) \quad \text{with} \quad \psi(x) = x\log x - x + 1.$$

In particular, for a.e. $t \in (0,T)$ it holds

$$\lim_{\varepsilon\to 0} \mathcal{F}^\varepsilon_{\mathrm{mic}}(m^\varepsilon(t)) = 0 \qquad \text{and} \qquad \lim_{\varepsilon\to 0} \mathcal{F}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon) = E(\nu_t). \tag{2.32}$$

Remark 2.14. The statement (2.32) is not enough to ensure well-prepared initial data, since the statement only holds for a.e. $t \in [0,T]$. However, it suggests that the statement of Corollary 2.11 holds already under the assumption of *macroscopically well-prepared* initial data:

$$\lim_{\varepsilon\to 0} E(\nu_0^\varepsilon) = E(\nu_0). \tag{2.33}$$

The assumption (2.33) together with the tightness Assumption 2.7 are natural, since they are also a sufficient condition for establishing continuous dependency on the initial data for the limiting gradient flow (cf. Proposition 2.8).

Remark 2.15. It is possible to use a different rescaling of the Becker–Döring system with different assumptions on the coagulation and fragmentation rates to obtain the LSW equation in the limit (cf. [5, 12]). Recently, within this scaling regime a quasi steady approximation was used to derive a suitable boundary condition for the macroscopic limits (cf. [7]).

3. The LSW equation and its gradient structure

To make the above formal calculation from Section 2.2 rigorous, we introduce the concept of curves of finite action.

Definition 3.1 (Curves of finite action)**.** A weakly* continuous curve $[0,T] \ni t \mapsto \nu_t \in M$ is called a curve of finite action, if there exists a measurable vector field $[0,T] \ni t \mapsto w_t \in T^*_{\nu_t}M$ such that

$$A(\nu, w) := \int_0^T \int \lambda^\alpha |w_t|^2 \, \nu_t(\mathrm{d}\lambda) < \infty,$$

where the pair $(\nu, w) \in \mathrm{CE}_T$ solves the continuity equation

$$\partial_t \nu_t + \partial_\lambda(\lambda^\alpha w_t \nu_t) = 0 \quad \text{in} \quad C_c^\infty([0,T] \times \mathbb{R}_+)^*. \tag{3.1}$$

Remark 3.2 (Wasserstein distance with weighted mobility). We can define a transportation distance induced from the metric (2.20) as a Wasserstein-type distance defined via the Benamou–Brenier formula

$$W^2_{\mathrm{LSW}}(\nu^0, \nu^T) := \inf\left\{ \int_0^T A(\nu_t, w_t) \, \mathrm{d}t : (\nu, w) \in \mathrm{CE}_T(\nu^0, \nu^T) \right\}$$

and $(\nu, w) \in \mathrm{CE}_T(\nu^0, \nu^T)$ are solution to (3.1) with $\nu_0 = \nu^0$ and $\nu_T = \nu^T$. Note, that $\nu^0, \nu^T \in M$ are not probability measures, but have equal first moment. For the convergence statement, we do not need the induced distance and suspend further investigation for the moment.

Before formulating the compactness statement, we want to revise the definition of the dissipation (2.21) and generalize it to curves of finite action. The dissipation acts as a weak upper gradient. Hence, for a curve of finite action $[0,T] \ni t \mapsto \nu_t \in M$ and using the fact that $w_t \in T^*_{\nu_t}M$ for all $t \in [0,T]$ it formally follows

$$\begin{aligned} |E(\nu_T) - E(\nu_0)| &= \left| - \int_0^T \int q\lambda^{\alpha-\gamma} w_t \, \mathrm{d}\nu_t \, \mathrm{d}t \right| \le \int_0^T \lambda^\alpha \left| \int \left(u(t) - q\lambda^{-\gamma}\right) \, w_t \, \mathrm{d}\nu_t \right| \mathrm{d}t \\ &\le \int_0^T \left(\int \lambda^\alpha \left(u(\nu_t) - q\lambda^{-\gamma}\right)^2 \mathrm{d}\nu_t \right)^{\frac{1}{2}} (A(\nu_t, w_t))^{\frac{1}{2}} \, \mathrm{d}t, \end{aligned} \tag{3.2}$$

where $u(\nu_t)$ is an arbitrary function on M. The choice of $u(\nu_t)$ is fixed by a minimization in L^2. That is, we define the dissipation as the weighted L^2-minimal upper gradient for the energy. Before doing so, we need as an auxiliary result, that a finite dissipation implies the existence of the α-moment for a curve of finite action.

Lemma 3.3 (Moment estimate)**.** *Assume $\alpha \ge 1 - 3\gamma$. Let $(\nu, w) \in \mathrm{CE}_T$ be a curve of finite action in M such that*

$$\inf_{u \in L^2([0,T])} \int_0^T \int \lambda^\alpha \left(u(t) - q\lambda^{-\gamma}\right)^2 \mathrm{d}\nu_t \, \mathrm{d}t < \infty. \tag{3.3}$$

Then, it holds the moment estimate

$$\int_0^T \int \lambda^\alpha \, \mathrm{d}\nu_t \, \mathrm{d}t < \infty. \tag{3.4}$$

Proof. Let us define $D(\nu,u) = \int \lambda^\alpha (u - q\lambda^{-\gamma})^2 \,\mathrm{d}\nu$. We observe that for $\alpha \geq 1-\gamma$, there is nothing to show, since the bound follows by interpolation from $\sup_{t\in[0,T]} E(\nu_t) < \infty$ and $\int \lambda \,\mathrm{d}\nu_t = \bar\varrho$.

Therefore, assume now $\alpha \leq 1-\gamma$. Let us define $\eta(\lambda) := \lambda\chi_{[0,1]}(\lambda) + \chi_{(1,\infty)}(\lambda)$. Then, we can estimate with Cauchy–Schwarz for any $\kappa \in \mathbb{R}$

$$\begin{aligned}\int_0^T \left(\int \bigl(u(t) - q\lambda^{-\gamma}\bigr)\eta(\lambda)^\kappa \,\mathrm{d}\nu_t\right)^2 \mathrm{d}t &\leq \int_0^T D(\nu_t,u(t)) \int \eta(\lambda)^{2\kappa}\lambda^{-\alpha}\,\mathrm{d}\nu_t\,\mathrm{d}t \\ &\leq \int_0^T D(\nu_t,u(t))\,\mathrm{d}t \sup_{t\in[0,T]} \int \eta(\lambda)^{2\kappa}\lambda^{-\alpha}\,\mathrm{d}\nu_t.\end{aligned} \tag{3.5}$$

Since, $\sup_{t\in[0,T]} E(\nu_t) < \infty$ and $\int \lambda\,\mathrm{d}\nu_t = \bar\varrho$, we can use interpolation to bound the sup in t provided $2\kappa - \alpha \geq 1-\gamma$. On, the other hand, since $\int \lambda\,\mathrm{d}\nu_t = \bar\varrho$ for all $t \geq 0$, there exists a constant $\bar\varrho_T > 0$ for any $T > 0$ such that $\int \eta(\lambda)\,\mathrm{d}\nu_t \geq \bar\varrho_T$ (see also Lemma 4.8 for a similar argument). Therewith, we can estimate the left hand side of (3.5) from below in the case $\kappa = 1$ by using the Young inequality for some $0 < \tau < 1$

$$\begin{aligned}\int_0^T \left(\int \bigl(u(t) - q\lambda^{-\gamma}\bigr)\eta(\lambda)\,\mathrm{d}\nu_t\right)^2 \mathrm{d}t &\geq (1-\tau)\bar\varrho_T^2 \int_0^T u(t)^2\,\mathrm{d}t \\ &\quad - \left(\frac{1}{\tau} - 1\right)\int_0^T \bigl((1-\gamma)E(\nu_t)\bigr)^2\,\mathrm{d}t.\end{aligned}$$

Since, $E(\nu_t) \in L^\infty([0,T])$, we obtain the first a priori estimate

$$\int_0^T u(t)^2\,\mathrm{d}t \leq C_T \int_0^T D(\nu_t,u(t))\,\mathrm{d}t + C_T. \tag{3.6}$$

Another choice is $\kappa = 1-\gamma$ thanks to $\alpha \leq 1-\gamma$. Then, we estimate the left hand side of (3.5) by using again the Young inequality with $\tau \in (0,1)$ as follows

$$\begin{aligned}\int_0^T \left(\int \bigl(u(t) - q\lambda^{-\gamma}\bigr)\eta(\lambda)^{1-\gamma}\,\mathrm{d}\nu_t\right)^2 \mathrm{d}t &\geq (1-\tau)q \int \left(\int_0^1 \lambda^{1-2\gamma}\,\mathrm{d}\nu_t\right)^2 \mathrm{d}t \\ &\quad - \left(\frac{1}{\tau} - 1\right)\int_0^T u(t)^2\bigl((1-\gamma)E(\nu_t)\bigr)^2\,\mathrm{d}t.\end{aligned}$$

Since, we trivially have $\int_1^\infty \lambda^{1-2\gamma}\,\mathrm{d}\nu_t \leq \int \lambda\,\mathrm{d}\nu_t = \bar\varrho$, it follows by using the first a priori bound (3.6) and $E(\nu_t) \in L^\infty([0,T])$ the second a priori estimate

$$\int_0^T \left(\int \lambda^{1-2\gamma}\,\mathrm{d}\nu_t\right)^2 \mathrm{d}t \leq C_T \int_0^T D(\nu_t,u(t))\,\mathrm{d}t + C_T, \tag{3.7}$$

which shows (3.4) for $\alpha \geq 1-2\gamma$. Hence, we assume now $\alpha \leq 1-2\gamma$. Similarly to (3.5), we can now estimate by Cauchy–Schwarz for some $\tilde\kappa \in \mathbb{R}$

$$\begin{aligned}\int_0^T \left|\int \bigl(u(t) - q\lambda^{-\gamma}\bigr)\eta(\lambda)^{\tilde\kappa}\,\mathrm{d}\nu_t\right| \mathrm{d}t \leq \left(\int_0^T D(\nu_t,u(t))\,\mathrm{d}t\right)^{\frac12} &\times \\ \left(\int_0^T \int \eta(\lambda)^{2\tilde\kappa}\lambda^{-\alpha}\,\mathrm{d}\nu_t\,\mathrm{d}t\right)^{\frac12}&.\end{aligned} \tag{3.8}$$

The second factor is bounded for $2\tilde\kappa - \alpha \geq 1-2\gamma$ by (3.7). Hence, a possible choice is $\tilde\kappa = 1-2\gamma$ by the assumption $\alpha \leq 1-2\gamma$. Therewith, since $u \in L^2((0,T))$ and $\int \lambda^{1-2\gamma}\,\mathrm{d}\nu_t \in L^2((0,T))$, we conclude the estimate (3.4). □

The Lemma provides the crucial ingredient to conclude that the dissipation is well-defined and justifies the use of the weak formulation in the first step of (3.2).

Proposition 3.4. *Assume $\alpha \geq 1-3\gamma$. Let $(\nu, w) \in \mathrm{CE}_T$ be a curve of finite action in M such that (3.3) holds. Then the associated minimization problem has a unique solution $u \in L^2([0,T])$ such that*

$$\lambda \mapsto u(t) - q\lambda^{-\gamma} \in T^*_{\nu_t} M \qquad \text{for a.e. } t \in [0,T]. \tag{3.9}$$

Moreover, the associated functional defined for a.e. $t \in [0,T]$ by

$$D(\nu_t) := \int \lambda^\alpha \big(u(t) - q\lambda^{-\gamma}\big)^2 \,\mathrm{d}\nu_t \quad \text{with} \quad u(t) := \frac{q\int \lambda^{\alpha-\gamma}\,\mathrm{d}\nu_t}{\int \lambda^\alpha \,\mathrm{d}\nu_t}, \tag{3.10}$$

called dissipation, *is a* strong upper gradient *for the energy E. That is, it holds for any curve $(\nu, w) \in \mathrm{CE}_T$ of finite action*

$$|E(\nu_t) - E(\nu_s)| \leq \int_s^t \sqrt{D(\nu_r)}\,\sqrt{A(\nu_r, w_r)}\,\mathrm{d}r, \qquad \forall 0 \leq s < t \leq T. \tag{3.11}$$

Hereby, equality in (3.11) holds if and only if $w_t(\lambda) = \pm(u(t) - q\lambda^{-\gamma})$ for ν_t-a.e. $\lambda \in \mathbb{R}_+$.

Proof. In the first step, we show (3.9) and (3.10). Therefore, the first variation of the minimization problem (3.3) along some $s : \mathbb{R}_+ \to \mathbb{R}$ is given by

$$\int_0^T \int \big(u(t) - q\lambda^{-\gamma}\big)\lambda^\alpha \,\mathrm{d}\nu_t\, s(t)\,\mathrm{d}t = 0.$$

We show that is is well-defined by an estimate analog to (3.8)

$$\left|\int_0^T \lambda^\alpha \big(u(t) - q\lambda^{-\gamma}\big)\,\mathrm{d}\nu_t\; s(t)\,\mathrm{d}t\right| \leq \left(\int_0^T D(\nu_t, u(t))\,\mathrm{d}t\right)^{\frac{1}{2}} \times \left(\int_0^T s(t)^2 \int \lambda^\alpha \,\mathrm{d}\nu_t\,\mathrm{d}t\right)^{\frac{1}{2}},$$

which is bounded thanks to the estimate (3.4) for $s \in L^\infty((0,T))$. In addition the a prior estimate (3.6) shows that minimizer is actually in $L^2((0,T))$ and hence satisfying the Euler-Lagrange equation $\int (u(t) - q\lambda^{-\gamma})\lambda^\alpha \,\mathrm{d}\nu_t = 0$ for a.e. $t \in [0,T]$, which is nothing else than (3.9) also showing (3.10).

It is left to show, that $D(\nu_t)$ is a strong upper gradient for the energy. Therefore, we fix a test function $\zeta \in C_c^\infty(\mathbb{R}_+)$ and calculate for a curve $(\nu, w) \in \mathrm{CE}_T$

$$\frac{\mathrm{d}}{\mathrm{d}t}\frac{q}{1-\gamma}\int \lambda^{1-\gamma}\zeta \,\mathrm{d}\nu_t = q\int \lambda^{\alpha-\gamma}\zeta\, w_t \,\mathrm{d}\nu_t + \frac{q}{1-\gamma}\int \lambda^{1+\alpha-\gamma}\zeta'\, w_t \,\mathrm{d}\nu_t =: \mathrm{I} + \mathrm{II}\,.$$

Using the fact that $w_t \in T^*_{\nu_t} M$, we can smuggle in $u(t)$ and apply Cauchy–Schwarz to the first term I, to obtain

$$\mathrm{I} \leq A(\nu_t, w_t)^{\frac{1}{2}} \left(\int \lambda^\alpha (u - q\lambda^{-\gamma}\zeta)^2 \,\mathrm{d}\nu_t\right)^{\frac{1}{2}},$$

Hereby, equality holds if and only if $w_t = \pm w_t^\zeta$ with $w_t^\zeta := u - q\lambda^{-\gamma}\zeta$. Hence, by choosing ζ_n converging to 1 from below the result (3.11) follows by integration in time and dominated convergence, provided the term II vanishes. By an additional approximation step, we can justify to choose the sequence $\zeta_n(\lambda) = n\lambda\chi_{[0,1/n)} + \chi_{[1/n,\infty)}$ and therewith, we can estimate II by

$$\mathrm{II} \leq \frac{1}{1-\gamma}\left(\int_0^{\frac{1}{n}} \lambda^\alpha |w_t|^2 \,\mathrm{d}\nu_t\right)^{\frac{1}{2}} \left(\int_0^{\frac{1}{n}} \lambda^\alpha \big|u - q\lambda^{-\gamma}\lambda\zeta_n'\big|^2 \,\mathrm{d}\nu_t\right)^{\frac{1}{2}}.$$

Since, we can assume the r.h.s. of (3.11) to be finite, we can conclude again by dominated convergence, that $\mathrm{II} \to 0$ as $n \to \infty$, which finishes the proof. □

Lemma 3.5 (Tightness is preserved by curves of finite action)**.** *Let ν_0 satisfy the tightness Assumption 2.7. Then for any $T > 0$ and any curve $(\nu, w) \in \mathrm{CE}_T$ of finite action ν_t satisfies the tightness Assumption 2.7 uniformly for all $t \in [0, T]$.*

Proof. Fix a test function $\eta_{r,R} \in C_c^\infty(\mathbb{R}_+, [0,1])$ such that $\eta_{r,R}(s) = 0$ for $s < r/2$ and $s > 2R$, $\eta_{r,R}(s) = 1$ for $r \le s \le R$, $|\eta'_{r,R}(s)| \le C/r$ for $r/2 \le s < r$ as well as $|\eta'_{r,R}(s)| \le C/R$ for $R \le s < 2R$. Therewith, we can estimate for a curve of finite action $(\nu, w) \in \mathrm{CE}_T$

$$
\begin{aligned}
\left| \frac{\mathrm{d}}{\mathrm{d}t} \int \lambda \eta(\lambda) \,\mathrm{d}\nu_t \right| &\le \int \lambda^\alpha \, \eta \, |w_t| \,\mathrm{d}\nu_t + \int \lambda^{1+\alpha} \, |\eta'| \, |w_t| \,\mathrm{d}\nu_t \\
&\le \frac{C}{r^{\frac{1-\alpha}{2}}} \int_{\frac{r}{2}}^\infty \lambda^{\frac{1+\alpha}{2}} |w_t| \,\mathrm{d}\nu_t + \frac{C}{r^{\frac{1-\alpha}{2}}} \int_{\frac{r}{2}}^r \lambda^{\frac{1+\alpha}{2}} |w_t| \,\mathrm{d}\nu_t \\
&\quad + \frac{C}{R^{\frac{1-\alpha}{2}}} \int_R^{2R} \lambda^{\frac{1+\alpha}{2}} |w_t| \,\mathrm{d}\nu_t \\
&\le C \left(\frac{1}{r^{\frac{1-\alpha}{2}}} + \frac{1}{R^{\frac{1-\alpha}{2}}} \right) A(\nu_t, w_t)^{\frac{1}{2}} \left(\int \lambda \,\mathrm{d}\nu_t \right)^{\frac{1}{2}}.
\end{aligned}
$$

By an integration in time, letting $R \to \infty$ and using the assumption of finite action, we obtain for all $t \in [0, T]$ the estimate

$$
\int_r^\infty \lambda \,\mathrm{d}\nu_t \le \int_{\frac{r}{2}}^\infty \lambda \,\mathrm{d}\nu_0 + \frac{C\sqrt{T}}{r^{\frac{1-\alpha}{2}}}.
$$

Since ν_0 is tight, we obtain that also ν_t is tight. □

Proposition 3.6 (Compactness of curves of finite action)**.** *Assume $\alpha \ge 1 - 3\gamma$ and let $(\nu^n, w^n) \in \mathrm{CE}_T$ for $n \in \mathbb{N}$ be a family of solutions to the continuity equation with uniformly bounded action and dissipation such that $\{\nu_0^n\}_{n \in \mathbb{N}}$ satisfies the tightness Assumption 2.7. Then, there exists a subsequence and a couple $(\nu, w) \in \mathrm{CE}_T$, such that*

$$
\begin{aligned}
\nu_t^n &\overset{*}{\rightharpoonup} \nu_t \qquad \text{in } C_c^0(\mathbb{R}_+)^* \quad \forall t \in [0, T], \\
w^n \nu^n &\overset{*}{\rightharpoonup} w\nu \qquad \text{in } C_c^0([0, T] \times \mathbb{R}_+)^*.
\end{aligned} \tag{3.12}
$$

In addition, the action and dissipation satisfy the lim inf *estimates*

$$
\int_0^T A(\nu_t, w_t) \,\mathrm{d}t \le \liminf_{n \to \infty} \int_0^T A(\nu_t^n, w_t^n) \,\mathrm{d}t \tag{3.13}
$$

$$
\int_0^T D(\nu_t) \,\mathrm{d}t \le \liminf_{n \to \infty} \int_0^T D(\nu_t^n) \,\mathrm{d}t. \tag{3.14}
$$

Proof. For any $\xi \in C_c^1(\mathbb{R}_+)$ and $0 \le t_1 < t_2 \le T$ holds

$$
\left| \int \zeta \,\mathrm{d}\nu_{t_2}^n - \int \zeta \,\mathrm{d}\nu_{t_1}^n \right| \le \sup_{\lambda > 0} \frac{|\partial_\lambda \zeta|}{\lambda^{\frac{1-\alpha}{2}}} |t_2 - t_1|^{\frac{1}{2}} \left(\int_{t_1}^{t_2} A(\nu_t^n, w_t^n) \,\mathrm{d}t \right)^{\frac{1}{2}},
$$

which shows (3.12) and the weak* continuity of ν_t. Moreover, it holds for $\kappa \in \mathbb{R}$ and $\zeta \in C_c^0([0, T] \times \mathbb{R}_+)$

$$
\begin{aligned}
\int_0^T \int \zeta(t, \lambda) \lambda^{\kappa + \alpha} |w_t^n| \,\mathrm{d}\nu_t^n \,\mathrm{d}t \le &\left(\int_0^T A(\nu_t^n, w_t^n) \,\mathrm{d}t \right)^{\frac{1}{2}} \times \\
&\left(\int_0^T \int \zeta(t, \lambda)^2 \lambda^{2\kappa + \alpha} \,\mathrm{d}\nu_t \,\mathrm{d}t \right)^{\frac{1}{2}}.
\end{aligned} \tag{3.15}
$$

By lower semi-continuity follows $E(\nu_t) < \infty$ and by the tightness Lemma 3.5 follows the preservation of total mass $\int \lambda \, d\nu_t = \int \lambda \, d\nu_0 = \int \lambda \, d\nu_0^n < \infty$. Hence, $\nu_t \in M$ for all $t \in [0,T]$. Therewith, it follows by interpolation, that the second term in (3.15) is finite for $1-\gamma \leq 2\kappa + \alpha \leq 1$. Hence, there exists $\mu \in C_c^0([0,T]\times\mathbb{R}_+)$ such that $w^n\nu^n \overset{*}{\rightharpoonup} \mu$ and the pair (ν,μ) satisfies $\partial_t\nu_t + \partial_\lambda\mu_t = 0$ in $C_c^\infty([0,T]\times\mathbb{R}_+)^*$. Since (ν^n, w^n) is a curve of finite action, we find a subsequence such that

$$\lim_{n\to\infty}\int_0^T A(\nu_t^n, w_t^n)\, dt = A^* := \liminf_{n\to\infty}\int_0^T\int_0^T A(\nu_t^n, w_t^n)\, dt.$$

Hence, we get the estimate with $\kappa = (1-\alpha)/2$

$$\int_0^T\int \zeta(t,\lambda)\lambda^{\frac{1-\alpha}{2}}\mu_t(d\lambda)\, dt \leq \left(A^*\int_0^T\int \zeta(t,\lambda)^2\lambda\, d\nu_t\, dt\right)^{\frac12}. \tag{3.16}$$

Now, we can apply the Riesz representation theorem to find $v \in L^2(\lambda\, d\nu_t dt)$ such that

$$\int_0^T\int \zeta(t,\lambda)\lambda^{\frac{1-\alpha}{2}}\mu_t(d\lambda)\, dt = \int_0^T\int \zeta(t,\lambda)\lambda v(t,\lambda)\, \nu_t(d\lambda)\, dt.$$

Setting $\tilde\zeta(t,\lambda) = \lambda^{\frac{1-\alpha}{2}}\zeta(t,\lambda)$ and $w_t(\lambda) = \lambda^{\frac{1-\alpha}{2}}v(t,\lambda)$, we get that $\mu_t(d\lambda) = v(t,\lambda)\nu_t(d\lambda)$. Moreover, since $w \in L^2(\lambda^\alpha\, d\nu_t\, dt)$ it is of finite action. Moreover, by approximating $\zeta(t,\lambda) = \frac{w_t(\lambda)}{z_s\lambda^{\frac{1-\alpha}{2}}}$ it follows from (3.16) the lower semi-continuity of the action (3.13).

Finally, (3.14) follows by noting that $D(\nu_t^n) = A(\nu_t^n, u(\nu_t^n) - q\lambda^{-\gamma})$, which is well-defined by (3.9). □

The formulation of the LSW gradient flow as curves of minimal action, reads now in analog to Proposition 2.5

Proposition 3.7 (LSW equation as curves of maximal slope)**.** *Let $\alpha \geq 1-3\gamma$. For $(\nu,w)\in \mathrm{CE}_T$ with finite action holds*

$$J(\nu) := E(\nu_T) - E(\nu_0) + \frac12\int_0^T D(\nu_t)\, dt + \frac12\int_0^T A(\nu_t, w_t)\, dt \geq 0. \tag{3.17}$$

Moreover, equality holds if and only if ν_t is a solution to the LSW equation.

Proof. We can assume that the dissipation $\int_0^T D(\nu_t)\, dt$ is bounded, because else there is nothing to show. Then, we can use the strong upper gradient property of the dissipation (3.11) after an application of the Young inequality to arrive at

$$\frac{d}{dt}E(\nu_t) \geq -\frac12 D(\nu_t) - \frac12 A(\nu_t, w_t).$$

An integration of the above estimate shows the nonnegativity of J in (3.17). The equality case follows from the equality case in (3.11) for a.e. $t\in[0,T]$ by choosing $w_t(\lambda) = u(\nu_t) - ql^{-\gamma}$. Then, by weak* continuity of $t\mapsto\nu_t$ follows the result for all $t\in[0,T]$.

Now, for a curve $(\nu,w)\in\mathrm{CE}_T$ with $J(\nu)=0$ follows by Proposition (3.4) and (3.11) the identity

$$-\int \lambda^\alpha q\lambda^{-\gamma}w_t\, d\nu_t = \sqrt{A(\nu_t,w_t)D(\nu_t)} = A(\nu_t,w_t) = D(\nu,w_t).$$

Since $w_t \in T^*_{\nu_t}M$, it follows that $w_t = u(t) - q\lambda^{-\gamma}$ for ν_t almost every $\lambda\in\mathbb{R}_+$. Hence, the continuity equation (3.1) takes the form

$$\partial_t\nu_t + \partial_\lambda\big(\lambda^\alpha(u(t)-q\lambda^{-\gamma})\nu_t\big) = 0 \quad \text{in} \quad C_c^\infty([0,T]\times\mathbb{R}_+)^*,$$

which is nothing else than a weak solution to the LSW equation. □

The compactness statement Proposition 3.6 with the variational characterization of solutions of the LSW equation from Proposition 3.7 is the essential tool to show the continuous dependence.

Proof of Proposition 2.8. By the compactness statement Proposition (3.6) follows that there exists a couple $(\nu_t, w_t) \in \mathrm{CE}_T$ being the weak* limit of $(\nu_t^\varepsilon, w_t^\varepsilon) \in \mathrm{CE}_T$ and satisfying the two lim inf estimates (3.13) and (3.14). By lower semi-continuity of the energy and the assumption (2.23) follows $0 = \liminf_{\varepsilon\to 0} J(\nu^\varepsilon) \geq J(\nu) \geq 0$ and hence $J(\nu) = 0$, which proves the claim. □

Remark 3.8. The compactness statement in Proposition 3.6 is also a tool to proof existence of solution to the LSW equation by the particle method (cf. [17, 18]). Therefore, the initial distribution is approximated in the weak* sense by a discrete sum of Dirac deltas. Solutions for such data are determined by solving the finite system of ordinary differential equations determined by (2.18) for each particle. Then the compactness statement allows to pass to the limit in the particle number and existence for measure valued initial distributions is obtained.

4. Proof of main results

4.1. **A priori estimates for the Becker–Döring gradient structure.** In this section, we consider the Becker–Döring equation. Let us recall the necessary ingredients for its gradient structure: The stoichiometric coefficients x and y are given by

$$x_l^r = \delta_{1,l} + \delta_{r,l} \qquad \text{and} \qquad y_l^r = \delta_{r+1,l}.$$

Therewith, the conserved quantities orthogonal to the stoichiometric subspace $\mathcal{S}$ (2.5) are the total numbers of monomers among all cluster. Hence the manifold or set of states is given by

$$\mathcal{M} = \mathcal{M}_{\varrho_0} = \left\{ n \in \mathbb{R}^{\mathbb{N}} : \sum_{l=1}^{\infty} l n_l = \varrho_0 \right\}.$$

The forward and backward reaction rates satisfy the Assumption 1.1, which is identical to the choice in [16, (1.5),(1.6)]. Then, the reversible equilibrium distribution ω with parameter $z \in (0, z_s]$ (corresponding to the conserved quantity) is given by

$$\omega_l(z) = z^l Q_l \qquad \text{with} \qquad Q_1 = 1 \quad \text{and} \quad Q_l = \prod_{j=1}^{l-1} \frac{a_j}{b_j}.$$

Note, that the radius of convergence for $z \mapsto \sum_{l=1}^{\infty} l\omega_l(z)$ is z_s and $\sum_{l=1}^{\infty} l\omega_l =: \varrho_s < \infty$ (cf. Lemma 4.1 below). Hence, the equilibrium state $\omega_l := \omega_l(z_s)$ is the one with largest total mass ϱ_s. We will work in the excess mass regime and any state will have total mass larger than ϱ_s to which there doesn't exist an according equilibrium state with the same total mass. The free energy is always the relative entropy with respect to $\omega = \omega(z_s)$, if not stated explicitly.

Lemma 4.1. *Under Assumption 1.1, there exists a constant $\mathcal{F}_0$ such that for any $l \geq 2$*

$$Q_l = \frac{1}{l^\alpha z_s^{l-1}} \exp\left(\left(\mathcal{F}_0 - \frac{q}{z_s}(1-\gamma) l^{1-\gamma} + \frac{q^2}{2 z_s^2 (1-2\gamma)} \left(l^{1-2\gamma} - 1 \right) \right) \left(1 + O(l^{-\gamma}) \right) \right), \tag{4.1}$$

where $\frac{l^{1-2\gamma}-1}{1-2\gamma} := \log l$ for $\gamma = \frac{1}{2}$.

The proof relies on elementary estimates and is included for convenience in Appendix B. The expansion of the rates allows us to easily conclude the expansion of the free energy $\mathcal{F}$.

Lemma 4.2 (Expansion of free energy)**.** *Let $n \in \mathcal{M}$ be given such that $\mathcal{F}(n) < \infty$ as defined in (2.4), then there exists $\sigma > 0$ such that for any $l_0 \geq 2$*

$$\mathcal{F}_{l_0}(n) = \mathcal{F}_{l_0}^{\mathrm{LSW}}(n)\big(1 + O(l_0^{-\sigma}) + O(l_0^{\gamma}\omega_{l_0})\big), \tag{4.2}$$

where $\mathcal{F}_{l_0}^{\mathrm{LSW}}$ is defined by

$$\mathcal{F}_{l_0}^{\mathrm{LSW}}(n) := \frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} n_l.$$

Proof. We expand the function ψ in the definition of $\mathcal{F}_{l_0}$

$$\mathcal{F}_{l_0}(n) = \sum_{l=l_0}^{\infty} \left(n_l \log \frac{1}{z_s^l Q_l} + n_l(\log n_l - 1) + \omega_l \right)$$

We estimate the first sum using the asymptotic expansion (4.1)

$$\begin{aligned}
\sum_{l=l_0}^{\infty} \left(n_l \log \frac{1}{z_s^l Q_l} \right) &= \frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} n_l + O\left(\sum_{l=l_0}^{\infty} l^{1-2\gamma} n_l \right) \\
&\quad + \sum_{l=l_0}^{\infty} l^{1-\gamma} n_l \frac{\log l^{\alpha}}{l^{1-\gamma}} - \sum_{l=l_0}^{\infty} \frac{l^{1-\gamma}}{l^{1-\gamma}} n_l \big(\log z_s + \mathcal{F}_0\big(1 + O(l^{-\gamma})\big)\big) \\
&= \left(\frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} n_l \right) \Big(1 + O\big(l_0^{-\gamma} \log l_0\big) + O\big(l_0^{-(1-\gamma)}\big) \Big),
\end{aligned}$$

Likewise, we note that for any $\beta \in (0,1)$ exists $C_\beta > 0$ such that for $x > 0$

$$|\min\{x(\log x - 1), 0\}| \leq C_\beta x^{\beta}.$$

Therewith and with the Hölder inequality, we can estimate

$$\begin{aligned}
\sum_{l=l_0}^{\infty} |\min\{n_l(\log n_l - 1), 0\}| &\leq C_\beta \sum_{l=l_0}^{\infty} n_l^{\beta} \\
&\leq C_\beta \left(\sum_{l=l_0}^{\infty} l^{1-\gamma} n_l \right)^{\beta} \left(\sum_{l=l_0}^{\infty} \frac{1}{l^{\frac{\beta}{1-\beta}(1-\gamma)}} \right)^{1-\beta}.
\end{aligned}$$

Now, we can choose β such that $\frac{\beta}{1-\beta}(1-\gamma) = \kappa > 1$ and $\beta < 1$ leading to the estimate

$$\sum_{l=l_0}^{\infty} |\min\{n_l(\log n_l - 1), 0\}| \leq C_\beta \left(\beta \sum_{l=l_0}^{\infty} l^{1-\gamma} n_l + 1 - \beta \right) O(l_0^{-(\kappa-1)(1-\beta)}).$$

The last term evaluates with the help of (4.1) to

$$\begin{aligned}
\sum_{l=l_0}^{\infty} \omega_l &\leq \sum_{l=l_0}^{\infty} \frac{z_s}{l^{\alpha}} \exp\left(\left(\mathcal{F}_0 - \frac{q}{z_s(1-\gamma)} l^{1-\gamma} \right) \big(1 + O(l^{-\gamma})\big) \right) \\
&\leq C \int_{l_0}^{\infty} \exp\left(-\frac{q}{z_s(1-\gamma)} l^{1-\gamma} \right) \mathrm{d}l \leq C\, l_0^{\gamma} \exp\left(-\frac{q}{z_s(1-\gamma)} l_0^{1-\gamma} \right)
\end{aligned}$$

Therefore, a combination of all the estimates leads to the result. □

Moreover, we need a Czisar-Pinsker inequality for the free energy, which was already a crucial ingredient in [16]

Proposition 4.3 (Czisar-Pinsker inequality [16, Lemma 2.1, 2.2]). *For $n \in \mathcal{M}$ and any small $\eta > 0$ and any $p < \infty$ and any $l_0 \geq 2$ holds*

$$\sum_{l=1}^{\infty} l^{1-\gamma} |n_l - \omega_l| \leq C\sqrt{\mathcal{F}(n)} \tag{4.3}$$

$$\left| \sum_{l=l_0}^{\infty} l n_l - (\varrho - \varrho_s) \right| \leq C l_0^{\gamma} \sqrt{\mathcal{F}(n)} + C_p l_0^{-p}. \tag{4.4}$$

For the next Lemmata, we make statements on curves of finite action to deduce certain compactness, which we later need for passing to the limit. These Lemmata are the analog of [16, Lemma 2.3 and 2.4], but we have to proof them for curves of finite action instead of solutions to the Becker–Döring equation.

Lemma 4.4 (A priori estimates for curves of finite action). *Let $(n, \phi) \in \mathcal{CE}_T$ be a curve of finite action and $\eta \in L^2(0,T)$, then it holds*

$$\int_0^T \eta(t) \sum_{l=l_0}^{\infty} l^{\kappa} k^l \hat{n}_l^{\omega}(t) |\nabla_l \phi(t)| \,\mathrm{d}t \leq C \left(\sup_{t\in[0,T]} \mathcal{F}_{l_0}^{\mathrm{LSW}}(n(t)) \right)^{\frac{1-\alpha-2\kappa}{2\gamma}} \tag{4.5}$$
$$\times \int_0^T |\eta(t)| \sqrt{\mathcal{A}_{\mathrm{mac}}(n(t), \phi(t))} \,\mathrm{d}t,$$

for any $\kappa \in \left[\frac{1-\alpha-\gamma}{2}, \frac{1-\alpha}{2}\right]$ with $\mathcal{A}_{\mathrm{mac}}$ the action as defined in (2.9) restricted to $l \geq l_0$. Hereby, $\nabla_l \phi := (y^l - x^l) \cdot \phi = \phi_{l+1} - \phi_l - \phi_1$ and $\hat{n}_l^{\omega}$ is defined by

$$\hat{n}_l^{\omega} := \Lambda\left(\frac{n_1 n_l}{\omega_1 \omega_l}, \frac{n_{l+1}}{\omega_{l+1}} \right).$$

Moreover, it also holds the estimate

$$\int_0^T \eta(t) \sum_{l=l_0}^{\infty} l^{\kappa} |a_l n_1(t) n_l(t) - b_{l+1} n_{l+1}(t)| \leq C \left(\sup_{t\in[0,T]} \mathcal{F}_{l_0}^{\mathrm{LSW}}(n(t)) \right)^{\frac{1-\alpha-2\kappa}{2\gamma}} \tag{4.6}$$
$$\times \int_0^T |\eta(t)| \sqrt{\mathcal{D}_{\mathrm{mac}}(n(t))} \,\mathrm{d}t,$$

where again $\mathcal{D}_{\mathrm{mac}}$ is defined as in (1.6) restricted to $l \geq l_0$.

Proof. We estimate using the Cauchy–Schwarz inequality

$$\sum_{l=l_0}^{\infty} l^{\kappa} k_l \hat{n}_l^{\omega} |\nabla_l \phi(t)| \leq \left(\sum_{l=l_0}^{\infty} k_l \hat{n}_l^{\omega} |\nabla_l \phi(t)|^2 \right)^{\frac{1}{2}} \left(\sum_{l=l_0}^{\infty} l^{2\kappa} k_l \hat{n}_l^{\omega} \right)^{\frac{1}{2}}.$$

Now, using that fact that

$$k_l \hat{n}_l^{\omega} = \Lambda(a_l n_1 n_l, b_{l+1} n_{l+1}) = \Lambda\big(\lambda^{\alpha} n_1 n_l, (\lambda+1)^{\alpha} (z_s + q(l+1)^{-\gamma}) n_{l+1}\big),$$

the estimate $\frac{(\lambda+1)^{\alpha}}{\lambda^{\alpha}} \leq 1 + \frac{\alpha}{\lambda}$ and from (4.3) the bound $|n_1 - z_s| \leq C\sqrt{\mathcal{F}(n)}$, it follows

$$\sum_{l=l_0}^{\infty} l^{2\kappa} k_l \hat{n}_l^{\omega} \leq 2\Big(z_s + \max\Big\{ C\sqrt{\mathcal{F}(n)}, q l_0^{-\gamma} \Big\} \Big) \sum_{l=l_0}^{\infty} l^{\alpha+2\kappa} n_l.$$

Now, we use the Hölder inequality to interpolate

$$\sum_{l=l_0}^{\infty} l^{\alpha+2\kappa} n_l \leq \left(\sum_{l=l_0}^{\infty} l n_l \right)^{\frac{\alpha+2\kappa+\gamma-1}{\gamma}} \left(\sum_{l=l_0}^{\infty} l^{1-\gamma} n_l \right)^{\frac{1-\alpha-2\kappa}{\gamma}} \leq C \left(\mathcal{F}_{l_0}^{\mathrm{LSW}}(n(t)) \right)^{\frac{1-\alpha-2\kappa}{\gamma}}$$

by assuming $1-\alpha-\gamma \le 2\kappa \le 1-\alpha$. The estimate (4.6) follows from (4.5) by noting that with the choice $\phi^*(t) = D\mathcal{F}(n(t)) = \left(\log \frac{n_l(t)}{\omega_l}\right)_{l\ge 1}$ holds

$$k^l \hat{n}_l^\omega(t)|\nabla_l \phi^*(t)| = a_l n_1(t) n_l(t) - b_{l+1} n_{l+1}(t)$$

and $\mathcal{A}_{\mathrm{mac}}(n(t),\phi^*(t)) = \mathcal{D}_{\mathrm{mac}}(n(t))$. □

The last a priori estimate deals with tightness and how tightness is preserved for curves of finite action.

Assumption 4.5 (Tightness)**.** *$n \in \mathcal{M}$ is called tight provided that*

$$\sum_{l=R}^{\infty} l n_l \to 0 \qquad \text{as } R \to \infty. \tag{4.7}$$

Lemma 4.6 (Tightness is preserved for curves of finite action)**.** *Let $(n,\phi) \in \mathcal{CE}_T$ be a curve of finite action and let $n(0)$ satisfy the tightness assumption (4.7), then for any $t \in [0,T]$ also $n(t)$ satisfies the tightness assumption (4.7).*

Proof. The proof is similar to Lemma 3.5, where the same result is proven for the LSW gradient structure. Let $1 \ll M \ll N$ and let $\eta \in C^1(\mathbb{R})$ be a cut off function such that $\eta(l) = 0$ for $l \le \frac{M}{2}$ and $l \ge 2N$, $\eta(l) = 1$ for $M \le l \le N$ and such that $\eta'(l) \le \frac{C}{M}$ for $\frac{M}{2} \le l \le M$ and $|\eta'(l)| \le \frac{C}{N}$ for $N \le l \le 2N$. Moreover, we define $\mathcal{N}_l := l\eta(l)$ and assume $M > 2$ such that $\eta(1) = 0$. Therewith, it follows

$$\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{N}\cdot n(t) &= \mathcal{N}\cdot\partial_t n(t) = \mathcal{N}\cdot\mathcal{K}(n)\phi \\
&= \left(\sum_{l=1}^{\infty} \eta(l+1)\, k_l\, \widehat{n}_l^\omega(t)\, \nabla_l\phi(t) + \sum_{l=1}^{\infty} k_l\, \widehat{n}_l^\omega(t)\, l\, \nabla_l\eta\, \nabla_l\phi(t)\right) \\
&\le C\left(\frac{1}{M^{\frac{1-\alpha}{2}}}\sum_{l=M/2}^{\infty} l^{\frac{1-\alpha}{2}} k_l \widehat{n}_l^\omega(t)|\nabla_l\phi(t)| \right.\\
&\qquad \left. + \left(\frac{1}{M}M^{1-\frac{1-\alpha}{2}} + \frac{1}{N}N^{1-\frac{1-\alpha}{2}}\right)\sum_{l=M/2}^{\infty} l^{\frac{1-\alpha}{2}} k_l \widehat{n}_l^\omega(t)|\nabla_l\phi(t)|\right) \\
&\le C\left(\frac{1}{M^{\frac{1-\alpha}{2}}} + \frac{1}{N^{\frac{1-\alpha}{2}}}\right)\sum_{l=M/2}^{\infty} l^{\frac{1-\alpha}{2}} k_l \widehat{n}_l^\omega(t)|\nabla_l\phi(t)|.
\end{aligned}$$

Integrating over time and using (4.5) leads to

$$\sum_{l=M}^{N} l n_l(t) \le \sum_{l=M/2}^{2N} l n_l(0) + C\left(\frac{1}{M^{\frac{1-\alpha}{2}}} + \frac{1}{N^{\frac{1-\alpha}{2}}}\right)\int_0^t \sqrt{\mathcal{A}(n(t),\phi(t))}\,\mathrm{d}t.$$

Now, using the fact that $t \mapsto n(t)$ is a curve of finite action and letting $N \to \infty$, we obtain

$$\sum_{l=M}^{\infty} l n_l(t) \le \sum_{l=M/2}^{\infty} l n_l(0) + \frac{Ct^{\frac{1}{2}}}{M^{\frac{1-\alpha}{2}}},$$

which finishes the proof since $n(0)$ satisfies the tightness assumption 4.5. □

4.2. **Passage to the limit: Proof of Theorem 2.10.** To pass to the limit in the discrete continuity equation, we define the flux density measure for a fixed covector ϕ and rescaled one $w^\varepsilon(\varepsilon l) = \varepsilon^{-\gamma}\nabla_l \phi$ (cf. (2.14)) by

$$\begin{aligned} \mu^\varepsilon(\mathrm{d}\lambda) &:= \frac{1}{\varepsilon^{1-\alpha}} \sum_{l\geq l_0} \delta_{\varepsilon l}(\mathrm{d}\lambda) k_l\, \widehat{n^\varepsilon}^\omega \nabla_l \phi^\varepsilon \\ &= \frac{1}{\varepsilon^{1-\alpha+\gamma}} \sum_{l\geq l_0} \delta_{\varepsilon l}(\mathrm{d}\lambda) l^\alpha \Lambda\big(n_1^\varepsilon n_l^\varepsilon, (z_s + q(l+1)^{-\gamma}) n_{l+1}^\varepsilon\big) w^\varepsilon(\lambda). \end{aligned} \tag{4.8}$$

and the dissipation flux density measure

$$\hat\mu^\varepsilon(\mathrm{d}\lambda) := \frac{1}{\varepsilon^{1-\alpha+\gamma}} \sum_{l\geq l_0} \delta_{\varepsilon l}(\mathrm{d}\lambda)(a_l n_1(t) n_l(t) - b_{l+1} n_{l+1}(t)) \tag{4.9}$$

Let us note, that with the above definitions for $l \geq l_0$ and $\lambda = \varepsilon l$ holds

$$\dot n_l^\varepsilon(t) - \frac{1}{\varepsilon^{1-\alpha+\gamma}} (\mathcal{K}[n]\phi)_l = \partial_t \nu_t^\varepsilon(\lambda) + \partial_\lambda^\varepsilon \mu_t^\varepsilon(\lambda) = 0, \tag{4.10}$$

where

$$\partial_\lambda^\varepsilon \mu_t^\varepsilon(\lambda) := \frac{\mu_t^\varepsilon(\lambda+\varepsilon) - \mu_t^\varepsilon(\lambda)}{\varepsilon}.$$

Therewith, let us summarize the a priori estimates found in Section 4.1 and rewrite them in rescaled variables.

Proposition 4.7 (Rescaled a priori estimates)**.** *With x from* (2.13) *holds*

i) The rescaled free energy satisfies

$$C \geq \mathcal{F}^\varepsilon(n^\varepsilon) \geq \mathcal{F}^\varepsilon_{\mathrm{mac}}(\nu^\varepsilon) = \frac{1}{z_s} E(\nu^\varepsilon)(1 + O(\varepsilon^{x\sigma})) \tag{4.11}$$

ii) The total excess mass satisfies

$$\left| \int \lambda \nu^\varepsilon(\mathrm{d}\lambda) - (\varrho_0 - \varrho_s) \right| \leq C \varepsilon^{\gamma\left(\frac12 - x\right)} \sqrt{\mathcal{F}^\varepsilon_{\mathrm{mac}}(\nu^\varepsilon)}. \tag{4.12}$$

iii) Let $[0,T] \ni t \mapsto \nu_t^\varepsilon \in \mathcal{M}^\varepsilon$ be a rescaled curve of finite action and $\eta \in L^2((0,T))$, then

$$\int_0^T \eta(t) \int \lambda^\kappa |\mu_t^\varepsilon(\mathrm{d}\lambda)|\,\mathrm{d}t \leq C \left(\sup_{t\in[0,T]} \mathcal{F}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon)\right)^{\frac{1-\alpha-2\kappa}{2\gamma}} \int_0^T |\eta(t)| \sqrt{\mathcal{A}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon, w_t^\varepsilon)}\,\mathrm{d}t, \tag{4.13}$$

$$\int_0^T \eta(t) \int \lambda^\kappa |\hat\mu_t^\varepsilon(\mathrm{d}\lambda)|\,\mathrm{d}t \leq C \left(\sup_{t\in[0,T]} \mathcal{F}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon)\right)^{\frac{1-\alpha-2\kappa}{2\gamma}} \int_0^T |\eta(t)| \sqrt{\mathcal{D}^\varepsilon_{\mathrm{mac}}(\nu_t^\varepsilon)}\,\mathrm{d}t, \tag{4.14}$$

for $\kappa \in \left[\frac{1-\alpha-\gamma}{2}, \frac{1-\alpha}{2}\right]$.

iv) If ν_0^ε satisfies Assumption 2.7 and $[0,T] \ni t \mapsto \nu_t^\varepsilon \in \mathcal{M}^\varepsilon$ are rescaled curves of finite action, then for all $t \in [0,T]$

$$\sup_{M\to\infty} \int_M^\infty \lambda\, \nu_t^\varepsilon(\mathrm{d}\lambda) \to 0 \qquad \textit{as } M \to \infty \quad \textit{uniformly in } \varepsilon. \tag{4.15}$$

The above results enable us to conclude the lim inf estimates and proof Theorem 2.10.

Proof of Theorem 2.10. Step 1: Convergence of ν^ε. For $\zeta \in C_c^1(\mathbb{R}_+)$ and $0 \le t_1 < t_2 \le T$, we calculate using the discrete continuity equation in the form (4.10)

$$\begin{aligned}\left|\int \zeta \,\mathrm{d}\nu_{t_1}^\varepsilon - \int \zeta \,\mathrm{d}\nu_{t_1}^\varepsilon\right| &= \left|\int_{t_1}^{t_2} \int \partial_\lambda^\varepsilon \zeta(\lambda) \mu_t^\varepsilon(\mathrm{d}\lambda)\,\mathrm{d}t\right| \\ &\le \sup_{\lambda\in\mathbb{R}_+} \frac{|\partial_\lambda^\varepsilon \zeta(\lambda)|}{\lambda^{\frac{1-\alpha}{2}}} \int_{t_1}^{t_2} \int \lambda^{\frac{1-\alpha}{2}} |\mu_t^\varepsilon(\mathrm{d}\lambda)|\,\mathrm{d}t \\ &\overset{(4.13)}{\le} C \sup_{\lambda\in\mathbb{R}_+} \frac{|\partial_\lambda^\varepsilon \zeta(\lambda)|}{\lambda^{\frac{1-\alpha}{2}}} \int_{t_1}^{t_2} \sqrt{\mathcal{A}_{\mathrm{mac}}^\varepsilon(\nu_t^\varepsilon, w_t^\varepsilon)}\,\mathrm{d}t \\ &\le C \sup_{\lambda\in\mathbb{R}_+} \frac{|\zeta'(\lambda)|}{\lambda^{\frac{1-\alpha}{2}}} \sqrt{|t_1 - t_2|}.\end{aligned}$$

This estimate together with the bound (4.11) imply via Arzelà-Ascoli the weak* convergence towards a weakly* continuous map $t \mapsto \nu_t$. Moreover, the a priori bounds (4.11), (4.12) and (4.15) imply that $\int \zeta \,\mathrm{d}\nu^\varepsilon \to \int \zeta \,\mathrm{d}\nu$ holds for $\zeta \in C^0(\mathbb{R}_+)$ satisfying

$$\limsup_{\lambda\to\infty} \frac{|\zeta(\lambda)|}{\lambda} < \infty \qquad \text{and} \qquad \lim_{\lambda\to 0} \frac{|\zeta(\lambda)|}{\lambda^{1-\gamma}} = 0,$$

which implies that the excess mass is preserved

$$\int \lambda \,\mathrm{d}\nu_t = \varrho_0 - \varrho_s \quad \Rightarrow \quad \nu_t \in M, \qquad \text{for all } t \in [0,T].$$

Moreover, the bounds (4.11) and (4.12) also imply by weak lower semi-continuity the estimate (2.27) and especially that $\sup_{t\in[0,T]} E(\nu_t) < \infty$. *Step 2: Convergence of μ^ε.* The a priori estimate (4.13) implies the existence of a measure $\mu \in C_c^0([0,T]\times\mathbb{R}_+)^*$ such that up to subsequences

$$\int\int \zeta(t,\lambda)\,\mathrm{d}\mu_t^\varepsilon \;\mathrm{d}t \to \int\int \zeta(t,\lambda)\,\mathrm{d}\mu \qquad \text{for all } \zeta \in C_c^0([0,T]\times\mathbb{R}_+). \tag{4.16}$$

Now, we show the limiting measure is of the form $\mu(\mathrm{d}t,\mathrm{d}\lambda) = \lambda^\alpha w_t(\lambda)\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t$ for some vector field w_t with finite action. Therefore, we remind at the definition of μ^ε (4.8) and $\mathcal{A}_{\mathrm{mac}}^\varepsilon$ (2.15) to estimate

$$\int\int \zeta(t,\lambda)\lambda^{\frac{1-\alpha}{2}} \mu_t^\varepsilon(\mathrm{d}\lambda)\,\mathrm{d}t \le \left(\int_0^T \mathcal{A}_{\mathrm{mac}}^\varepsilon(\nu_t^\varepsilon, w_t^\varepsilon)\,\mathrm{d}t\right)^{\frac{1}{2}} \times \left(\int_0^T \sum_{l\ge l_0} \zeta^2(t,\varepsilon l)\, l^{1-\alpha} k_l\, \widehat{n^\varepsilon}_l^\omega(t)\,\mathrm{d}t\right)^{\frac{1}{2}}. \tag{4.17}$$

The second term on the right hand side can be bounded by using the one-homogeneity and concavity of $(a,b)\mapsto \Lambda(a,b)$

$$\begin{aligned}&\sum_{l\ge l_0} \zeta^2(t,\varepsilon l)\, l^{1-\alpha} k_l\, \widehat{n^\varepsilon}_l^\omega(t) \\ &\quad\le \sum_{l\ge l_0} \zeta^2(t,\varepsilon l)\, l\, \Lambda\big(n_1^\varepsilon(t) n_l^\varepsilon(t), (z_s + q(l+1)^{-\gamma}) n_{l+1}^\varepsilon(t)\big) \\ &\quad\le \Lambda\left(n_1^\varepsilon(t) \sum_{l\ge l_0} \zeta^2(t,\varepsilon l)\, l\, n_l^\varepsilon(t), \big(z_s + q l_0^{-\gamma}\big) \sum_{l\ge l_0} \zeta^2(t,\varepsilon l)\, l\, n_{l+1}^\varepsilon(t)\right) \\ &\quad\le (z_s + o(1)) \sum_{l\ge l_0} \zeta^2(t,\varepsilon l)\, l\, n_l^\varepsilon(t),\end{aligned}$$

where we used in the last estimate that $l_0 = \varepsilon^{-x}$, $|n_1^\varepsilon - z_s| \leq C\sqrt{\mathcal{F}(n)} \leq C\varepsilon^{\frac{\gamma}{2}}$ by (4.3) and the fact that ζ is uniformly continuous. Since $t \mapsto n^\varepsilon(t)$ is a curve of finite action and by the convergence of the total mass, it follows that the right hand of (4.17) is finite. Hence, we can pass to the limit $\varepsilon \to 0$ in (4.17) by the same argument as in (4.16). It follows for a subsequence which attains

$$\int_0^T \mathcal{A}^\varepsilon_{\mathrm{mac}}(\nu^\varepsilon, w^\varepsilon)\,\mathrm{d}t \to A^* := \liminf_{\varepsilon\to 0} \int_0^T \mathcal{A}^\varepsilon_{\mathrm{mac}}(\nu^\varepsilon, w^\varepsilon)\,\mathrm{d}t.$$

the estimate

$$\int\int \zeta(t,\lambda)\lambda^{\frac{1-\alpha}{2}}\mu_t(\mathrm{d}\lambda)\,\mathrm{d}t \leq \left(A^* z_s \int\int \zeta^2(t,\lambda)\lambda\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t\right)^{\frac{1}{2}},$$

with $\mu_t(\mathrm{d}\lambda)$ denoting the disintegration of μ in t. Hence, by the Riesz representation theorem there exists a function $v \in L^2(\lambda\,\mathrm{d}\nu_t\,\mathrm{d}t)$ such that

$$\int\int \zeta(t,\lambda)\lambda^{\frac{1-\alpha}{2}}\mu_t(\mathrm{d}\lambda)\,\mathrm{d}t = \int\int \zeta(t,\lambda)\,\lambda\, v(t,\lambda)\,\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t \tag{4.18}$$

and it holds the estimate

$$\int\int \zeta(t,\lambda)\,\lambda\, v(t,\lambda)\,\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t \leq \left(A^* z_s \int\int \zeta^2(t,\lambda)\lambda\,\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t\right)^{\frac{1}{2}}. \tag{4.19}$$

Setting $\tilde\zeta(t,\lambda) = \lambda^{\frac{1-\alpha}{2}}\zeta(t,\lambda)$ and $w_t(\lambda)\lambda^{\frac{1-\alpha}{2}}v(t,\lambda)$ the representation (4.18) is of the desired form (2.25). Moreover, since $w \in L^2(\lambda^\alpha\,\mathrm{d}\nu_t\,\mathrm{d}t)$ it is of finite action. Moreover, by approximating $\zeta(t,\lambda) = \frac{w_t(\lambda)}{z_s\lambda^{\frac{1-\alpha}{2}}}$ it follows from (4.19) the lower semi-continuity of the action (2.28). *Step 3: Convergence of the dissipation $\mathcal{D}^\varepsilon$.* We observe that $\mathcal{D}^\varepsilon_{\mathrm{mac}}(\nu^\varepsilon_t) = \mathcal{A}^\varepsilon_{\mathrm{mac}}(\nu^\varepsilon_t, \tilde w^\varepsilon)$, where $\tilde w^\varepsilon$ is the special vector field given by $-\nabla_l D\mathcal{F}^\varepsilon(\nu^\varepsilon_t)$, i.e. for all l

$$\varepsilon^\gamma \tilde w^\varepsilon_t(\varepsilon l) = \log\frac{n_1^\varepsilon n_l^\varepsilon}{\omega_1\omega_l} - \log\frac{n_{l+1}^\varepsilon}{\omega_{l+1}}.$$

Therefore, we can apply the same arguments of step 2, but now to the dissipation flux density $\hat\mu^\varepsilon$ defined in (4.9) and use the a priori estimate (4.14) to deduce the lim inf estimate

$$\liminf_{\varepsilon\to 0}\int_0^T \mathcal{D}^\varepsilon(\nu_t^\varepsilon)\,\mathrm{d}t \geq \frac{1}{z_s}\int_0^T A(\nu_t,\tilde w_t)\,\mathrm{d}t = \frac{1}{z_s}\int_0^T\int \lambda^\alpha|\tilde w_t|^2\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t,$$

for some $\tilde w \in C_c^0([0,T]\times\mathbb{R}_+)^*$. It, is left to show that $h^\varepsilon(t) \rightharpoonup h(t)$ in $L^2((0,T))$ and $\tilde w_t$ is of the form $h(t) - q/\lambda^\gamma$ wit $h \in L^2([0,T])$, however this statement follows exactly along the lines of [16, Lemma 2.6]. The final result (2.29) follows now by the definition of $D(\nu_t)$ as the infimum over all such $h \in L^2([0,T])$ from Lemma 3.4).

Step 4: Continuity equation holds. Finally, choosing a subsequence such that both convergences (2.24) and (2.25) holds for a test function $\zeta \in C_c^\infty([0,T]\times\mathbb{R})$, we can pass to the limit in the weak form of the discrete continuity equation (4.10)

$$\begin{array}{ccc}
\displaystyle\int_0^T\int \partial_t\zeta(t,\lambda)\nu_t^\varepsilon(\mathrm{d}\lambda)\,\mathrm{d}t & + \displaystyle\int\int \partial_\lambda^\varepsilon\zeta(t,\lambda)\mu_t^\varepsilon(\mathrm{d}\lambda)\,\mathrm{d}t & = 0 \\
\downarrow\ \varepsilon\to 0 & \downarrow\varepsilon\to 0 & \\
\displaystyle\int_0^T\int \partial_t\zeta(t,\lambda)\,\nu_t(\mathrm{d}\lambda)\,\mathrm{d}t & + \displaystyle\int\int \partial_\lambda\zeta(t,\lambda)\,\mu_t(\mathrm{d}\lambda)\,\mathrm{d}t & = 0,
\end{array}$$

which shows $(\nu, w) \in \mathrm{CE}_T$. □

4.3. **Quasistationary expansion: Proof of Theorem 2.13.** The proofs of Proposition 2.12 and Theorem 2.13 consists in several steps, which are formulated in the following Lemmata. In the proofs of this section, C is a generic constant, which is assumed to be independent of ε and only depending on the parameters inside of the rates from Assumption 1.1.

Lemma 4.8. *Assume that $\mathcal{F}^{\varepsilon}_{\mathrm{mac}}(\nu^{\varepsilon}) \leq C$, l_0 satisfies (2.13) and ν^{ε} satisfies the tightness assumption (4.15) uniformly in ε. Then for any $\kappa \in [0,1]$, there exists $c > 0$ such that*

$$\int \lambda^{\kappa} \,\mathrm{d}\nu^{\varepsilon} \geq c > 0 \qquad \text{uniformly in } \varepsilon > 0. \tag{4.20}$$

Proof of Lemma 4.8. The assumptions of the Lemma ensure the conservation of the excess mass (4.12). Together with the tightness assumption, we have for any $\kappa \leq 1$

$$\begin{aligned}\int \lambda^{\kappa}\, \nu^{\varepsilon}_t(\mathrm{d}\lambda) &\geq \int_0^M \lambda^{\kappa}\, \nu^{\varepsilon}_t(\mathrm{d}\lambda)\\ &\geq \frac{1}{M^{1-\kappa}} \int_0^M \lambda\, \nu^{\varepsilon}_t(\mathrm{d}\lambda)\\ &\geq \frac{1}{M^{1-\kappa}}\Big(\rho_0 - \rho_s - O_{\varepsilon}\Big(\varepsilon^{\gamma(1/2-x)}\Big) - o_M(1)\Big)\end{aligned}$$

Hence, we can choose M large enough but finite and ε small enough such that for some $c > 0$ the estimate (4.20) holds. □

Lemma 4.9. *For any $n \in \mathcal{M}$ holds*

$$(u-h)\log\left(\frac{1+u}{1+h}\right) \leq \frac{\mathcal{D}_{\mathrm{mac}}(n)}{A(z_s)}, \tag{4.21}$$

where $u := u(z_s)$,

$$h := \frac{n_1 - z_s}{z_s} \qquad \text{and} \qquad u(z) := \frac{B - A(z)}{A(z)},$$

and

$$A(z) := \sum_{l \geq l_0} a_l z n_l \qquad \text{and} \qquad B := \sum_{l\geq l_0} b_{l+1} n_{l+1}. \tag{4.22}$$

Moreover, if $\mathcal{F}(n) \leq C\varepsilon^{\gamma}$, l_0 satisfies (2.13) and ν^{ε} satisfies the tightness (4.15) assumption, then it holds for ε small enough and some $C > 0$ uniformly in ε the estimate (2.30) from Proposition 2.12.

Proof of Lemma 4.9. For the proof n is fixed such that $\mathcal{F}(n) \leq C\varepsilon^{\gamma}$. Then, we introduce two measures α and β on $\{l_0, l_0+1, \dots\}$

$$\forall l \geq l_0: \qquad \alpha_l(z) := a_l z n_l \qquad \text{and} \qquad \beta_l := b_{l+1} n_{l+1}$$

with partition sums $A(z)$ and B (4.22), respectively.

We introduce $\mathcal{D}_{\mathrm{mac},z}(n)$ the constant monomer density dissipation of the large clusters

$$\mathcal{D}_{\mathrm{mac},z}(n) := \sum_{l\geq l_0} (a_l z n_l - b_{l+1} n_{l+1}) \log \frac{a_l z n_l}{b_{l+1} n_{l+1}} \geq 0.$$

Note, that by this definition $\mathcal{D}_{\mathrm{mac}}(n) = \mathcal{D}_{\mathrm{mac},n_1}(n)$. By the definition (4.22), it follows $A(n_1) = (1+h)A(z_s)$ and the identity

$$u(n_1) = \frac{1}{1+h}(u(z_s) - h). \tag{4.23}$$

Now, rewrite and $\mathcal{D}_{\mathrm{mac},z}(n)$ and apply the Jensen inequality to the one-homogeneous convex function $a \mapsto a\log(1+a)$

$$\begin{aligned}\mathcal{D}_{\mathrm{mac},z}(n) &= \sum_{l\geq l_0} \alpha_l(z)\frac{\beta_l-\alpha_l(z)}{\alpha_l(z)}\log\biggl(1+\frac{\beta_l-\alpha_l(z)}{\alpha_l(z)}\biggr)\\ &\geq (B-A(z))\log\frac{B}{A(z)} = A(z)(u(z)\log(1+u(z))\end{aligned}$$

Hence, we obtain by setting $z=n_1$ and using (4.23), the estimate

$$A(z)(u(z)\log(1+u(z)) = A(z_s)(u-h)\log\biggl(1+\frac{u-h}{1+h}\biggr),$$

from where we conclude (4.21). By using the explicit expression of the rates (1.1) follows

$$\begin{aligned}2A(z_s)-B &= \sum_{l\geq l_0} z_s l^\alpha\biggl(1-\frac{q}{z_s l^\gamma}\biggr)n_l + z_s l_0^\alpha n_{l_0} + q l_0^{\alpha-\gamma} n_{l_0}\\ &\geq \biggl(1-\frac{q}{z_s l_0^\gamma}\biggr)A(z_s) \geq A(z_s)(1-O(\varepsilon^{x\gamma})),\end{aligned}$$

by the definition of l_0 (2.13). Hence, we have $B \leq A(z_s)(1+O(\varepsilon^{x\gamma}))$ and in particular with (4.23)

$$u(n_1) \leq \frac{u(z_s)}{1-|h|} + \frac{|h|}{1-|h|} \leq O(\varepsilon^{x\gamma}) + O\Bigl(\varepsilon^{\frac{\gamma}{2}}\Bigr) = O(\varepsilon^{x\gamma}), \tag{4.24}$$

where we used that with $\mathcal{F}(n) \leq C\varepsilon^\gamma$ also $|h| \leq C\varepsilon^{\frac{\gamma}{2}}$ from the estimate (4.3). The estimate (4.24) allows to linearize the bound (4.21) as follows

$$(u-h)\log\biggl(\frac{1+h}{1+u}\biggr) \geq \frac{(u-h)^2}{\max\{1+h,1+u\}} \geq \frac{(u-h)^2}{1+O(\varepsilon^{x\gamma})}.$$

Finally, to deduce the estimate (2.30), it is enough to rewrite it in rescaled variables and use the estimate (4.20) from Lemma 4.8

$$A = z_s\varepsilon^{1-\alpha}\int \lambda^\alpha \nu^\varepsilon(\mathrm{d}\lambda) \geq c\varepsilon^{1-\alpha} > 0.$$

□

The time-scale separation between the dynamic of the small clusters and the one of the large clusters is characterized by the following logarithmic Sobolev type inequality.

Proposition 4.10 (Microscopic energy-dissipation estimate)**.** *Let* $\omega_l(z) := z^l Q_l$. *Then for all* $n \in \mathbb{R}_+^{\mathbb{N}}$ *with* $\mathcal{F}_{\mathrm{mic}}(n) \leq C\varepsilon^\gamma$ *there exists* C_{EED} *independent of* ε *such that it holds*

$$\mathcal{H}_{\mathrm{mic}}(n \mid \omega(n_1)) \leq C_{\mathrm{EED}}\varepsilon^{-x(1-\alpha+\gamma)}\mathcal{D}_{\mathrm{mic}}(n), \tag{4.25}$$

where $\mathcal{H}_{\mathrm{mic}}$ *is the microscopic part of the relative entropy between* n *and* $\omega(n_1)$ *defined by*

$$\mathcal{H}_{\mathrm{mic}}(n \mid \omega(z)) := \sum_{l=1}^{l_0-1}\omega_l(z)\psi\biggl(\frac{n_l}{\omega_l(z)}\biggr) \qquad \text{with} \qquad \psi(a) = a\log a - a + 1.$$

Moreover, the estimates (2.31) *and* (2.32) *from Theorem 2.13 hold.*

Proof. We note, that the function $(a,b) \mapsto \varphi(a,b) := (a-b)(\log a - \log b)$ occurring in the definition of the dissipation is one-homogeneous. In addition, the following lower bound holds

$$(a-b)(\log a - \log b) \geq 4\Big(\sqrt{a} - \sqrt{b}\Big)^2.$$

Moreover, we remind that $\omega(z)$ satisfies the detailed balance condition $a_l\omega_1(z)\omega_l(z) = b_{l+1}\omega_{l+1}(z)$. Therewith, by choosing $z = n_1$, the dissipation can be rewritten and bounded from below by

$$\begin{aligned}\mathcal{D}(n) &= \sum_{l\geq 1} a_l n_1 \omega_l(n_1)\ \varphi\bigg(\frac{n_l}{\omega_l(n_1)}, \frac{n_{l+1}}{\omega_{l+1}(n_1)}\bigg) \\ &\geq 4\sum_{l\geq 1} a_l n_1 \omega_l(n_1)\Bigg(\sqrt{\frac{n_l}{\omega_l(n_1)}} - \sqrt{\frac{n_{l+1}}{\omega_{l+1}(n_1)}}\Bigg)^2 := \overline{\mathcal{D}}(n).\end{aligned}$$

Hence, instead of showing the estimate (4.25), it is sufficient to proof

$$\mathcal{H}_{\mathrm{mic}}(n \mid \omega(n_1)) \leq C_{\mathrm{EED}}\overline{\mathcal{D}}(n).$$

This inequality was investigated in [4]. To apply their result, we have to introduce the measures

$$l \in \{1,\dots,l_0-1\}: \quad \mu_l(z) := \frac{\omega_l(z)}{\sum_{l=1}^{l_0-1}\omega_l(z)} \quad \text{and} \quad \nu_l(z) := \frac{a_l\omega_l(z)}{\sum_{l=1}^{l_0-1}\omega_l(z)}.$$

If we can proof the following mixed logarithmic Sobolev inequality with the measures μ, ν

$$\mathrm{Ent}_\mu(f^2) := \sum_{l=1}^{l_0-1} \mu_l f_l^2 \log\frac{f_l^2}{\sum_{l=1}^{l_0-1} f_l^2\mu_l} \leq C_{\mathrm{LSI}}\sum_{l=1}^{l_0-1}\nu_l(f_l - f_{l+1})^2 \tag{4.26}$$

then [4, Proposition 3.2] implies

$$C_{\mathrm{EED}} \leq \frac{C_{\mathrm{LSI}}}{n_1^2}\left(n_1^2 + 2\left(\sum_{l=1}^{l_0-1} n_l\right)\left(\sum_{l=1}^{l_0-1}\omega_l(n_1)\right)\right). \tag{4.27}$$

To proof the mixed logarithmic Sobolev inequality (4.26), we use [4, Corollary 2.4 and Remark 2.5], from which we obtain the bound

$$C_{\mathrm{LSI}} \leq 480 \sup_{1<l<l_0} W_l(n_1)\log\bigg(\frac{W_1(n_1)}{W_l(n_1)}\bigg)V_l(n_1), \tag{4.28}$$

where

$$W_l(z) = \sum_{j=l}^{l_0-1}\omega_j(z) \quad \text{and} \quad V_l(z) = \sum_{j=1}^{l-1}\frac{1}{a_j z\omega_j(z)}.$$

We will establish the following estimates for $|z - z_s| \leq C\varepsilon^{\frac{\gamma}{2}}$ and some $C > 1$

$$\frac{1}{C}\omega_l(z) \leq W_l(z) \leq Cl^\gamma\omega_l(z) \tag{4.29}$$

$$V_l(z) \leq \frac{Cl^{\gamma-\alpha}}{\omega_l(z)} \tag{4.30}$$

We postpone the proof of the estimates and first show the final result. By a combination of (4.29) and (4.30) with (4.28), we obtain the estimate

$$C_{\mathrm{LSI}} \leq C\sup_{1<l<l_0} l^{2\gamma-\alpha}\log\bigg(\frac{C}{\omega_l(z)}\bigg).$$

By the expansion (4.1) follows

$$\frac{1}{\omega_l(z)} = z^l Q_l \le C l^\alpha \exp\biggl(\frac{q(1-\gamma)}{z_s} l^{1-\gamma} - l \log\frac{z}{z_s}\biggr) \le C \exp\bigl(C l^{1-\gamma}\bigr),$$

where we used that $l_0 \Bigl|\log\frac{z}{z_s}\Bigr|$ is uniformly bounded, because of $|z - z_s| \le C\varepsilon^{\frac{\gamma}{2}}$. Therewith, we obtain the upper bound

$$C_{\mathrm{LSI}} \le C \sup_{1<l<l_0} l^{1-\alpha+\gamma} \le C l_0^{1-\alpha+\gamma}.$$

A combination of this bound with (4.27) leads to the bound

$$C_{\mathrm{EED}} \le C\varepsilon^{-x(1-\alpha+\gamma)} \; \frac{n_1^2 + W_1(n_1)\sum_{l=1}^{l_0-1} n_l}{n_1^2}$$

The conclusion (4.25) follows now from (4.29), $|n_1 - z_s| \le C\varepsilon^{\frac{\gamma}{2}}$ and the estimate

$$\sum_{l=1}^{l_0-1} n_l \le \sum_{l\ge 1} l n_l = \varrho.$$

To proof the estimates (4.29) and (4.30), we first observe that by the assumption $|z - z_s| \le C\varepsilon^{\frac{\gamma}{2}}$, we have the comparison

$$\frac{\omega_l(z)}{\omega_l(z_s)} = \biggl(\frac{z}{z_s}\biggr)^l \le \Bigl(1 + C\varepsilon^{\frac{\gamma}{2}}\Bigr)^l \le \exp\Bigl(C\varepsilon^{\frac{\gamma}{2}} l_0\Bigr) \le C$$

by the choice of l_0 (2.13). In the complete analog way, we get $\frac{\omega_l(z)}{\omega_l(z_s)} \ge \frac{1}{C}$. Hence, it is enough to show (4.29) and (4.30) for $z = z_s$.

Therefore, we use the expansion (4.1) from Lemma 4.1 in the form: For some constant $C > 1$ and any $l \ge 1$ holds

$$\frac{1}{C l^\alpha z_s^{l-1}} \exp\biggl(-\frac{q}{z_s}(1-\gamma) l^{1-\gamma}\biggr) \le Q_l \le \frac{C}{l^\alpha z_s^{l-1}} \exp\biggl(-\frac{q}{z_s}(1-\gamma) l^{1-\gamma}\biggr).$$

The estimate (4.29) with $z = z_s$ is now proven

$$\begin{aligned} W_l(z) &\le \frac{C}{z_s^2} \sum_{j=l}^{l_0-1} \frac{1}{j^\alpha} \exp\biggl(-\frac{q}{z_s}(1-\gamma) j^{1-\gamma}\biggr) \le \frac{C}{l^\alpha} \sum_{j=l}^{l_0-1} \exp\biggl(-\frac{q}{z_s}(1-\gamma) j^{1-\gamma}\biggr) \\ &\le \frac{C}{l^\alpha} \int_l^{l_0-1} \exp\biggl(-\frac{q}{z_s}(1-\gamma) v^{1-\gamma}\biggr) \mathrm{d}v \le C l^{\gamma-\alpha} \int_{l^{1-\gamma}}^\infty \exp\biggl(-\frac{q}{z_s}(1-\gamma) v\biggr) \mathrm{d}v \\ &\le C l^\gamma \omega_l. \end{aligned}$$

The estimate (4.30) with $z = z_s$ follows similarly

$$\begin{aligned} V_l(z) &\le \frac{C}{z_s^2} \sum_{j=1}^{l-1} \exp\biggl(\frac{q}{z_s}(1-\gamma) l^{1-\gamma}\biggr) \le \frac{C}{z_s^2} \int_1^l \exp\biggl(\frac{q}{z_s}(1-\gamma) v^{1-\gamma}\biggr) \mathrm{d}v \\ &\le C \frac{(1-\gamma)}{z_s^2} l^\gamma \int_0^{l^{1-\gamma}} \exp\biggl(\frac{q}{z_s}(1-\gamma) v\biggr) \mathrm{d}v \le C \frac{l^{\gamma-\alpha}}{\omega_l}. \end{aligned}$$

Finally, the estimate (2.31) follows just by rescaling $\mathcal{D}_{\mathrm{mic}}$ and integrating the estimate along a curve of finite action. For the statement (2.32), we first observe that $\mathcal{F}_{\mathrm{mic}}(n) = \mathcal{H}_{\mathrm{mic}}(n \mid \omega)$ and get by writing $z = z_s e^h$

$$\begin{aligned}\mathcal{H}_{\mathrm{mic}}(n \mid \omega(z)) - \mathcal{F}_{\mathrm{mic}}(n) &= -\sum_{l=1}^{l_0-1} l n_l \log\frac{z}{z_s} - \sum_{l=1}^{l_0-1} \omega_l \left(1 - \left(\frac{z}{z_s}\right)^l\right) \\ &= h\left(\varrho_s - \sum_{l=1}^{l_0-1} l n_l + \sum_{l=1}^{l_0-1} l\omega_l \frac{e^{lh}-1}{lh} - \varrho_s\right)\end{aligned}$$

The first difference in the bracket can be bounded in terms of (4.4) from Lemma 4.3. The second difference can be explicitly expresses as follows

$$\begin{aligned}\left|\varrho_s - \sum_{l=1}^{l_0-1} l\omega_l \frac{e^{lh}-1}{lh}\right| &\le \sum_{l\ge l_0} l\omega_l + \sum_{l=1}^{l_0-1} l\omega_l \frac{e^{lh}-1-lh}{lh} \\ &\le C l_0^\gamma \omega_{l_0} + Ch\sum_{l=1}^{l_0-1} l^2\omega_l \le C(l_0^\gamma\omega_{l_0} + h),\end{aligned}$$

where, we used the upper in (4.29), which holds by the proof also with $l_0 = \infty$. Moreover, ω_l has arbitrary high moments following from the expansion (4.1). By the choice of l_0 (2.13) and again (4.1) follows that $l_0^\gamma \omega_{l_0} \le C\varepsilon^p$ for any $p > 0$. Hence, combining all these estimates and reminding that $\mathcal{F}_{\mathrm{mic}}(n) \le C\varepsilon^\gamma$, we get

$$|\mathcal{H}_{\mathrm{mic}}(n \mid \omega(z)) - \mathcal{F}_{\mathrm{mic}}(n)| \le C|h|\Big(\varepsilon^{-x\gamma}\sqrt{\mathcal{F}_{\mathrm{mic}}(n)} + \varepsilon^p + |h|\Big) \le C|h|(\varepsilon^\sigma + |h|),$$

where $\sigma = \gamma\big(\frac{1}{2} - x\big) > 0$ by (2.13). Hence, we can conclude for a curve of finite action

$$\begin{aligned}\int_0^T \mathcal{F}^\varepsilon_{\mathrm{mic}}(n(t))\,\mathrm{d}t &\le \varepsilon^{-\gamma}\int_0^T \mathcal{H}_{\mathrm{mic}}(n(t) \mid \omega(n_1(t)))\,\mathrm{d}t \\ &\quad + C\varepsilon^{-\gamma}\int_0^T \left|\log\left(\frac{n_1(t)}{z_s} - 1\right)\right|\left(\varepsilon^\sigma + \left|\log\left(\frac{n_1(t)}{z_s} - 1\right)\right|\right)\mathrm{d}t \\ &\le C\varepsilon^{(1-x)(1-\alpha+\gamma)}\int_0^T \mathcal{D}^\varepsilon_{\mathrm{mic}}(n(t))\,\mathrm{d}t \\ &\quad + C\varepsilon^{-\gamma}\int_0^T |n_1(t) - z_s|^2\,\mathrm{d}t + C\varepsilon^{\sigma-\gamma}\sqrt{T}\left(\int_0^T |n_1(t) - z_s|^2\,\mathrm{d}t\right)^{\frac{1}{2}}.\end{aligned}$$

The conclusion (2.32) follows by (2.26) from Theorem 2.10 . □

A. Gradient structures for coagulation and fragmentation models

A.1. Smoluchowski coagulation and fragmentation equation. The Becker–Döring clustering equation is itself just a special case in the more general class of Smoluchowski coagulation and fragmentation equations seen as the following family of chemical reactions

$$X_i + X_j \underset{b_{i,j}}{\overset{a_{i,j}}{\rightleftharpoons}} X_{i+j} \qquad \text{with} \qquad (i,j) \in \mathbb{N}\times\mathbb{N}.$$

Hence, the stoichiometric coefficients in (2.1) are given as $x_k^{(i,j)} = \delta_{i,k} + \delta_{j,k}$ and $y_k^{(i,j)} = \delta_{i+j,k}$. A gradient flow structure can be established under the assumption of detailed balance, which in this case does not necessarily hold: There exists a state $\omega \in \mathbb{R}^{\mathbb{N}}$ such that for $(i,j) \in \mathbb{N}\times\mathbb{N}$ holds

$$a_{i,j}\omega_i\omega_j = b_{i,j}\omega_{i+j}.$$

Then, the gradient flow (2.7) of the free energy as defined in (2.4) and $\mathcal{K}$ the Onsager operator defined in (2.6) is the Smoluchowski coagulation and fragmentation equation.

A.2. **Modified Becker–Döring system.** The modified Becker–Döring system was introduced by Dreyer and Duderstadt [8] claiming to be a more thermodynamic consistent model then the original one. The main feature is the introduction of a mixing entropy between the clusters. The free energy consists of the original free energy (2.4) plus a mixing entropy

$$\tilde{\mathcal{F}}(n) = \mathcal{F}(n) - N(n)(\log N(n) - 1) \quad \text{with} \quad N(n) = \sum_i n_i. \tag{A.1}$$

The most compact form of the free energy is $\mathcal{F}(n) = \sum_i \Bigl(n_i \log \frac{n_i}{\omega_i N(n)} + \omega_i \Bigr)$. Hence, the differential of the free energy differential is given by

$$D\tilde{\mathcal{F}}(n) = \left(\log \frac{n_1}{\omega_1 N(n)}, \dots, \log \frac{n_i}{\omega_i N(n)}, \dots \right). \tag{A.2}$$

The reaction is still of the same form like in the classical Becker–Döring system, i.e. we have $x_i^r = \delta_{i,1} + \delta_{i,r}$ and $y_i^r = \delta_{i,r+1}$ with detailed balance condition

$$a_r \omega_1 \omega_r = b_{r+1}\omega_{r+1} =: k^r,$$

which leads to the same possible equilibrium states $\omega_r(z) = z^r Q_r$ as for the usual Becker–Döring system (1.3). However, z has to be determined from the condition $N[\omega(z)] = N(n)$, for a detailed analysis of the equilibrium states, see [11].

Now, from (A.2), we further deduce

$$(x^r - y^r) D\mathcal{F}(n) = \log \frac{n_1 n_r}{\omega_1 \omega_r N(n)^2} - \log \frac{n_{r+1}}{\omega_{r+1} N(n)} = \log \frac{n_1 n_r}{\omega_1 \omega_r} - \log \frac{N(n) n_{r+1}}{\omega_{r+1}}.$$

The modified Onsager matrix is given as

$$\tilde{\mathcal{K}}(n) := \sum_r k^r \Lambda \left(\frac{n_1 n_r}{\omega_1 \omega_r}, \frac{N(n) n_{r+1}}{\omega_{r+1}} \right) (x^r - y^r) \otimes (x^r - y^r).$$

Therewith, the modified Becker–Döring equation is the gradient flow of the modified free energy (A.1)

$$\begin{aligned} \dot{n} = -\tilde{\mathcal{K}}(n) D\tilde{\mathcal{F}}(n) &= -\sum_r k^r \left(\frac{n_1 n_r}{\omega_1 \omega_r} - \frac{N(n) n_{r+1}}{\omega_{r+1}} \right) (x^r - y^r) \\ &= -\sum_r (a_r n_1 n_r - b_{r+1} N(n) n_{r+1})(x^r - y^r) = -\sum_r \tilde{J}_r (x^r - y^r). \end{aligned}$$

Hence, we get

$$\dot{n}_i = \tilde{J}_{i-1} - \tilde{J}_i \quad \text{with} \quad \tilde{J}_0 := -\sum_{r=1}^{\infty} \tilde{J}_r \quad \text{and} \quad \tilde{J}_r(n) := a_r n_1 n_r - b_{r+1} N(n) n_{r+1}.$$

B. Proof of Lemma 4.1

Proof of Lemma 4.1. We calculate using the definition (1.3) of Q_l

$$\log\bigl(l^\alpha z_s^{l-1} Q_l \bigr) = -\sum_{j=2}^{l} \log \left(1 + \frac{q}{z_s j^\gamma} \right)$$

The function $x \mapsto \log\left(1 + \frac{q}{z_s k^\gamma}\right)$ is positive, continuous and monotone decreasing to 0. Therefore, we can define the Euler number

$$C_1 := \lim_{l\to\infty} \left(\sum_{j=2}^{l} \log\left(1 + \frac{q}{z_s j^\gamma}\right) - \int_2^l \log\left(1 + \frac{q}{z_s x^\gamma}\right) \right).$$

Moreover, we get from the Euler-MacLaurin formula the estimate

$$\left| C_1 - \left(\sum_{j=2}^{l} \log\left(1 + \frac{q}{z_s j^\gamma}\right) - \int_2^l \log\left(1 + \frac{q}{z_s x^\gamma}\right) \right) \right| \leq \log\left(1 + \frac{q}{z_s l^\gamma}\right) \leq \frac{q}{z_s l^\gamma}.$$

The following bound

$$\frac{q}{z_s x^\gamma} - \frac{1}{2}\left(\frac{q}{z_s x^\gamma}\right)^2 \leq \log\left(1 + \frac{q}{z_s x^\gamma}\right) \leq \frac{q}{z_s x^\gamma} - \frac{1}{2}\left(\frac{q}{z_s x^\gamma}\right)^2 + \frac{1}{3}\left(\frac{q}{z_s x^\gamma}\right)^3$$

implies the estimate

$$0 \leq \frac{\int_2^l \log\left(1 + \frac{q}{z_s x^\gamma}\right) \mathrm{d}x}{\int_2^l \left(\frac{q}{z_s x^\gamma} - \frac{1}{2}\left(\frac{q}{z_s x^\gamma}\right)^2\right) \mathrm{d}x} - 1 \leq O(l^{-\gamma}),$$

hereby, we use the convention that $\frac{l^\kappa - 1}{\kappa} = \log l$ for $\kappa = 0$. Now, we can combine all the estimates to obtain

$$\begin{aligned}
\log\left(l^\alpha z_s^{l-1} Q_l\right) &= -\left(\sum_{j=2}^{l} \log\left(1 + \frac{q}{z_s j^\gamma}\right) - \int_2^l \log\left(1 + \frac{q}{z_s x^\gamma}\right) \mathrm{d}x \right) \\
&\quad - \left(1 + \frac{\int_2^l \log\left(1 + \frac{q}{z_s x^\gamma}\right) \mathrm{d}x}{\int_2^l \left(\frac{q}{z_s x^\gamma} - \frac{1}{2}\left(\frac{q}{z_s x^\gamma}\right)^2\right) \mathrm{d}x} - 1 \right) \int_2^l \left(\frac{q}{z_s x^\gamma} - \frac{1}{2}\left(\frac{q}{z_s x^\gamma}\right)^2 \right) \mathrm{d}x \\
&= \left(\mathcal{F}_0 - \frac{q}{z_s(1-\gamma)} l^{1-\gamma} + \frac{q^2}{2 z_s^2 (1 - 2\gamma)} l^{1-2\gamma} \right) \left(1 + O(l^{-\gamma})\right),
\end{aligned}$$

which concludes the proof by setting $\mathcal{F}_0 = \frac{q 2^{1-\gamma}}{z_s(1-\gamma)} - \frac{q^2 2^{1-2\gamma}}{2 z_s^2 (1-2\gamma)} - C_1$. □

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