

On a thermodynamically consistent modification of the Becker-Döring equations

M. Herrmann^{a,1}, M. Naldzhieva^{a,2}, B. Niethammer^{a,2,3}

^a*Humboldt Universität zu Berlin, Department of Mathematics,
Section for Applied Analysis,
Unter den Linden 6, D-10099 Berlin, Germany*

Abstract

Recently, Dreyer and Duderstadt have proposed a modification of the Becker–Döring cluster equations which now have a nonconvex Lyapunov function. We start with existence and uniqueness results for the modified equations. Next we derive an explicit criterion for the existence of equilibrium states and solve the minimization problem for the Lyapunov function. Finally, we discuss the long time behavior in the case that equilibrium solutions do exist.

Key words: Becker–Döring equations, coagulation and fragmentation, nonconvex Lyapunov function, existence of equilibrium, convergence to equilibrium

PACS: 05.45, 86.03, 82.60

1 Introduction

The Becker–Döring equations are an infinite set of kinetic equations that describe the dynamics of cluster formation in a system of identical particles. In this model, clusters can coagulate to form larger clusters or fragment to smaller ones. In what follows we describe clusters by their size $l \geq 2$, the number of particles in the cluster, and we denote by $z_l(t)$ the total number of l -clusters in the system at time t . Note that here we always assume that all l -clusters are uniformly distributed in the physical space. Moreover, the number of free atoms in the system is abbreviated with $z_1(t)$, so that the state

¹ E-Mail michaelherrmann@mathematik.hu-berlin.de

² supported by the DFG research center MATHEON, see www.matheon.de

³ E-Mail niethamm@mathematik.hu-berlin.de

of the complete system is given by a nonnegative sequence $z(t) = (z_l(t))_{l \in \mathbb{N}}$, where $0 \notin \mathbb{N}$.

The crucial assumption of Becker and Döring in [1] was that an l -cluster can change its size only by gaining a free atom (*coagulation*) to form an $(l + 1)$ -cluster, or loosing an atom (*fragmentation*) to form an $(l - 1)$ -cluster. In particular, for all $l \geq 2$ there are two typical transition rates, namely a *condensation rate* $\Gamma_l^C(t)$ and a *vaporization rate* $\Gamma_l^V(t)$ giving at time t the probability that a l -clusters gains or loses a 1-cluster, respectively. The net rate of conversion of l -clusters into $(l + 1)$ -clusters is denoted by $J_l(t)$. For $l \geq 2$ it reads

$$J_l(t) = \Gamma_l^C(t) z_l(t) - \Gamma_{l+1}^V(t) z_{l+1}(t), \quad (\text{BD1})$$

and the change of the total number of l -clusters for $l \geq 2$ is given by

$$\frac{d}{dt} z_l(t) = J_{l-1}(t) - J_l(t), \quad l \geq 2. \quad (\text{BD2})$$

To describe the change of $z_1(t)$, the number of free atoms, a different equation is needed because free particles are involved in all reactions in the system. Here we are only interested in the case that the total number of all atoms in the system is conserved, i.e. $\varrho(z(t)) = \text{const}$, where

$$\varrho(z) = \sum_{l=1}^{\infty} l z_l. \quad (1)$$

This constraint gives rise to $\frac{d}{dt} z_1(t) = -J_1(t) - \sum_{l=1}^{\infty} J_l(t)$, which can be expressed as follows

$$\frac{d}{dt} z_1(t) = J_0(t) - J_1(t), \quad J_0(t) = - \sum_{l=1}^{\infty} J_l(t). \quad (\text{BD3})$$

The system (BD1)–(BD3) was derived and investigated the first time by Frenkel in [2]. Clearly, the equations must be closed by some *constitutive assumptions* relating the rates $\Gamma_l^C(t)$ and $\Gamma_l^V(t)$ to the state $z(t)$ of the system.

In [3], Dreyer and Duderstadt give a historical overview on the Becker–Döring equations with mass conservation. As they point out, almost all of the literature is based on a misinterpretation of [1]: The quantities $z_l(t)$ are considered as the *volume densities* of l -clusters, and not as *numbers*. Clearly, this reinterpretation corresponds to the non-explicit assumption that the total volume of the system is conserved. Dreyer and Duderstadt criticize this standard interpretation and the resulting constitutive laws, and derive new closure laws from fundamental thermodynamic principles. Next we first summarize the standard model, and afterwards we describe the modified model in detail.

The standard model

In the standard model, see for instance [4,5,6], the dynamical equations (BD1)–(BD3) are closed by the following constitutive assumptions

$$\Gamma_l^C(t) = c_l z_1(t), \quad \Gamma_l^V(t) = d_l, \quad (\text{SM})$$

where c_l and d_l depend neither on the state z nor on the time t . In fact, this is reasonable if $z_1(t)$ is the volume density of free atoms. The coefficient c_l and d_l are then determined by some heuristic arguments. To give an example, a very common ansatz is

$$c_l = l^\alpha, \quad d_l = c_l \left(z_s + \frac{q}{l^\gamma} \right) \quad (2)$$

with $0 \leq \alpha < 1$, $z_s > 0$, $q > 0$, $\gamma < 1$, and

$\alpha = 1/3, \quad \gamma = 1/3$	for <i>diffusion controlled kinetics</i> in 3D,
$\alpha = 0, \quad \gamma = 1/2$	for <i>diffusion controlled kinetics</i> in 2D,
$\alpha = 2/3, \quad \gamma = 1/3$	for <i>interface reaction limited kinetics</i> in 3D,
$\alpha = 1/2, \quad \gamma = 1/2$	for <i>interface reaction limited kinetics</i> in 2D.

Within the standard model (BD1)–(BD3) with (SM) there exists a convex Lyapunov function L with

$$L(z) = \sum_{l=1}^{\infty} z_l \left(\ln \left(\frac{z_l}{Q_l} \right) - 1 \right), \quad Q_1 = 1, \quad Q_{l+1} = \prod_{n=1}^l \frac{c_n}{d_{n+1}}, \quad (3)$$

such that $L(z(t))$ decreases with time t for all solutions $z(t)$. An equilibrium state \bar{z} of the dynamics is a state for which all transfer rates J_l vanish. After some basic calculation we find that an equilibrium state \bar{z} and its density $\bar{\rho}$ are given by

$$\bar{z}_l = Q_l \mu^l, \quad \bar{\rho} = \sum_{l=1}^{\infty} l Q_l \mu^l. \quad (4)$$

With (2) it can be shown that the radius of convergence of the power series in (4) is z_s , and that for $\mu = z_s$ the series converges to $\rho_s = \sum_{l=1}^{\infty} l Q_l z_s^l$. In particular, ρ_s is the maximal value for the equilibrium density, and can be interpreted as saturation density. As a consequence, if the density ρ_0 of initial data exceeds ρ_s , for $t \rightarrow \infty$ the total mass of the system cannot be stored in a equilibrium solution, but the excess density $\rho_0 - \rho_s$ must be transferred into larger and larger clusters when time proceeds. However, this process is in general extremely slow if the excess density is small. This metastability has been

rigorously established in [6] for typical initial data. As a consequence, exact numerical simulations are difficult to perform and impossible to perform for small $\varrho_0 - \varrho_s$, see [7]. In addition, it has been established that the dynamics of large clusters after the metastable state can be described the classical Lifshitz-Slyozov-Wagner equation for coarsening [6,8,9].

The non-standard model of Dreyer and Duderstadt

Dreyer and Duderstadt [3] model the system of all clusters=*droplets* as mixture of different substances, where a droplet with l atoms is regarded as a particle of the substance l . To be more precise, Dreyer and Duderstadt introduce a maximal size l_{\max} for the droplets, and thus they consider a mixture of l_{\max} different substances. Since the maximal droplet size l_{\max} is usually very large, we are mainly interested in the limiting case $l_{\max} = \infty$.

The main advantage of this new approach is that thermodynamics is able to describe the equilibrium without any knowledge of the dynamical law. On the contrary, thermodynamics give some constraints for the dynamical law. The main ideas in [3] can be summarized as follows.

- (1) The Second Law of thermodynamics states that the *available free energy*, or *availability*, of the system becomes minimal in equilibrium. This follows from a careful evaluation of Clausius theorem, and reflects the assumption on the physical process.
- (2) The available free energy a_l for a single droplet with l atoms can be given explicitly in many situations, see for instance the examples below.
- (3) Thermodynamic mixture theory provides an explicit expression for the availability A of a many droplet system. In particular, with $a_1 = 0$ it follows that

$$A(z) = \sum_{l=1}^{\infty} a_l z_l + \sum_{l=1}^{\infty} z_l \ln \left(\frac{z_l}{N(z)} \right), \quad (5)$$

where $N(z)$ abbreviates the total number of all droplets, i.e.

$$N(z) = \sum_{l=1}^{\infty} z_l. \quad (6)$$

Note that the second sum in (5) takes care of the *entropy of mixing*.

- (4) The Second Law of thermodynamics requires that the availability A decreases with time for any real world process, and from this we obtain a consistency relation for the transition rates Γ_l^V and Γ_l^C , see below.

For convenience we set

$$q_l = \exp(-a_l) \quad \text{with} \quad q_1 = 1, \quad (7)$$

so that the availability A of the many-droplet system reads

$$A(z) = \sum_{l=1}^{\infty} z_l \ln \left(\frac{z_l}{q_l N(z)} \right) = \sum_{l=1}^{\infty} z_l \ln z_l - \sum_{l=1}^{\infty} z_l \ln q_l - N(z) \ln N(z). \quad (8)$$

Since the function $x \mapsto x \ln x$ is convex, we conclude that A is the sum of a convex, a linear and a concave functional. In particular, A is neither convex nor concave.

Next we evaluate the thermodynamic consistency relation mentioned above. A formal calculation yields

$$\begin{aligned} \frac{d}{dt} A(z) &= \left(\sum_{l=1}^{\infty} \left(1 + \ln \frac{z_l}{q_l} \right) \frac{d}{dt} z_l \right) - \left(1 + \ln N(z) \right) \frac{d}{dt} N(z) \\ &= \sum_{l=1}^{\infty} \ln \left(\frac{z_l}{q_l N(z)} \right) \frac{d}{dt} z_l = \sum_{l=1}^{\infty} \left(J_{l-1}(z) - J_l(z) \right) \ln \left(\frac{z_l}{q_l N(z)} \right) \\ &= \ln \left(\frac{z_1}{N(z)} \right) J_0(z) + \sum_{l=1}^{\infty} J_l(z) \left(\ln \left(\frac{z_{l+1}}{q_{l+1} N(z)} \right) - \ln \left(\frac{z_l}{q_l N(z)} \right) \right) \\ &= \ln \left(\frac{z_1}{N(z)} \right) \left(- \sum_{l=1}^{\infty} J_l(z) \right) + \sum_{l=1}^{\infty} J_l(z) \ln \left(\frac{q_l z_{l+1}}{q_{l+1} z_l} \right) \\ &= \sum_{l=1}^{\infty} J_l(z) \ln \left(\frac{q_l z_{l+1} N(z)}{q_{l+1} z_l z_1} \right) \\ &= \sum_{l=1}^{\infty} \left(\Gamma_l^C z_l - \Gamma_{l+1}^V z_{l+1} \right) \ln \left(\frac{q_l z_{l+1} N(z)}{q_{l+1} z_l z_1} \right) \end{aligned} \quad (9)$$

The Second Law of thermodynamics states that $\frac{d}{dt} A(z)$ is non-positive for all solutions of the Becker-Döring dynamics (BD1)–(BD3). Dreyer and Duderstadt satisfy this restriction by fixing the ratio between the transition rates via

$$\frac{\Gamma_{l+1}^V(t)}{\Gamma_l^C(t)} = \frac{q_l}{q_{l+1}} \frac{N(z(t))}{z_1(t)}, \quad (\text{NSM})$$

so that the net rates $J_l(t)$ for $l \geq 1$ read

$$J_l(t) = \Gamma_l^C(t) \left(z_l(t) - \frac{N(z(t))}{z_1(t)} \frac{q_l z_{l+1}(t)}{q_{l+1}} \right). \quad (10)$$

With (NSM) the production of availability becomes

$$\begin{aligned} \frac{d}{dt} A(z) &= \sum_{l=1}^{\infty} \Gamma_l^C \left(z_l - \frac{N(z)}{z_1} \frac{q_l}{q_{l+1}} z_{l+1} \right) \ln \left(\frac{q_l z_{l+1} N(z)}{q_{l+1} z_l z_1} \right) \\ &= \sum_{l=1}^{\infty} \Gamma_l^C (z_l - w_l) (\ln w_l - \ln z_l) \leq 0, \end{aligned} \quad (11)$$

with $w_l = N(z) z_{l+1} q_l / (q_{l+1} z_1)$. In particular, the availability A is a nonconvex Lyapunov function for the dynamical system (BD1)–(BD3) with (NSM).

In [3], Dreyer and Duderstadt derive the availability A for two important examples. As mentioned above, they always consider a system which contains only a single droplet with l atoms, and derive explicit expression for the availability a_l . The availability A of the many-droplet system is given by (5).

Example 1 corresponds to a simple vapor-liquid system, in which a single gaseous droplet with l atoms is included in a liquid matrix, both made from the same chemical substance as for instance water. The result is

$$a_0 = 1, \quad a_l = -\delta l + \gamma l^{\frac{2}{3}} \quad \text{for } l > 1. \quad (12)$$

where δ and γ are positive constants.

Example 2 is more complicated, and describes a single liquid droplet contained in a crystalline solid, where both are a binary mixture of Gallium and Arsenic. Moreover, the solid is surrounded by an inert gas with prescribed pressure. The resulting expressions for the availability show that a_l growth with l for large l , and this gives rise to the following simplified ansatz

$$a_l = +\beta l \quad \text{for } l \gg 1, \quad (13)$$

where β is a positive constant. We will show in Section 3 that both examples differ in the set of possible equilibrium states.

Although thermodynamics give a constraint for the dynamical law, we are free to choose the transition rates $\Gamma_l^C(t)$. In what follows we always assume that

$$\Gamma_l^C(t) = z_1(t) \gamma_l, \quad (14)$$

where γ_l is constant. We mention that other choices of the time dependence of $\Gamma_l^C(t)$ may be reasonable, which, however, change only the time scale of the evolution. Finally, we obtain the following system of equations

$$\frac{d}{dt} z_l(t) = J_{l-1}(z(t)) - J_l(z(t)) \quad \text{for } l \geq 1, \quad (\text{MBD1})$$

$$J_0(z) = - \sum_{l=1}^{\infty} J_l(z), \quad (\text{MBD2})$$

$$J_l(z) = \gamma_l \left(z_1 z_l - N(z) \frac{q_l}{q_{l+1}} z_{l+1} \right) \quad \text{for } l \geq 1. \quad (\text{MBD3})$$

In what follows we will refer to this system as the *modified Becker-Döring equations*. Moreover, we always assume

$$0 < R := \lim_{l \rightarrow \infty} \frac{q_l}{q_{l+1}} < \infty, \quad (\text{A1})$$

as well as

$$\lim_{l \rightarrow \infty} \frac{\gamma_l}{l} = 0. \quad (\text{A2})$$

Note that (A1) implies the identity $1/R = \lim_{l \rightarrow \infty} q_l^{1/l}$.

Aims and results

This paper is organized as follows. In Section 2 we give a brief survey on existence and uniqueness results for the modified equations. We will skip some technical details, because in this part we mainly adapt methods which are well established for the standard model.

In Section 3 we investigate equilibrium states for the dynamical equations. Our first result is a necessary and sufficient condition (EQ) for the existence of such equilibrium states. Since this condition depends only on some properties of the sequence $(a_l)_{l \in \mathbb{N}}$, there is no upper bound for the mass of an equilibrium state. In other words, (EQ) implies that for all $\bar{\varrho} > 0$ there exists a unique and nonnegative equilibrium state \bar{z} with $\varrho(\bar{z}) = \bar{\varrho}$. Moreover, in Section 3 we study the minimization problem $A(z) \rightarrow \min$ under the constraint $\varrho(z) = \bar{\varrho}$, where $\bar{\varrho} > 0$ is fixed, and we prove the following two statements. 1. If (EQ) is satisfied, then the equilibrium state with mass $\bar{\varrho}$ is a minimizer. 2. In the case that (EQ) is violated there is no minimizer at all, but the infimum is $\bar{\varrho} \ln R$.

Section 4 is devoted to the limit $t \rightarrow \infty$, where the main problem is the following. Although the mass is conserved for finite times, see Section 2, some amount of mass may disappear in the limit $t \rightarrow \infty$. At first we show that for $t \rightarrow \infty$ the state $z(t)$ converges (in some weak sense) either to an equilibrium state with positive mass or to 0. Second, we state and prove an sufficient condition for that the mass remains conserved for $t \rightarrow \infty$. Finally, we identify several cases, and prove for most of them that either all mass is conserved or all mass disappears.

2 Existence and Uniqueness

Our main goal within this section is to prove the global existence of non-negative, *weak* solutions for the initial value problem of (MBD1)–(MBD3), see Theorem 4 below. Furthermore, we will explain how uniqueness results can be derived. For these reasons we fix some nonnegative initial data \tilde{z} with $\varrho_0 := \varrho(\tilde{z}) > 0$ and $\tilde{z}_l \geq 0$ for all $l \in \mathbb{N}$, and for simplicity we assume $\tilde{z}_1 > 0$. We seek solutions $t \mapsto z(t)$ of the Becker-Döring equations in the

space $C([0, \infty); X)$, where the *state space* X is given by

$$X = \left\{ z = (z_l)_{l \in \mathbb{N}} : \|z\|_X < \infty \right\}, \quad \|z\|_X = \sum_{l=1}^{\infty} l |z_l|, \quad (15)$$

Since we are only interested in solutions of the Becker-Döring equations which are positive or at least nonnegative, we introduce the cones X_{0+} and X_+ of all nonnegative and strictly positive, respectively, elements of X , i.e.

$$X_{0+} = \left\{ z \in X : z_l \geq 0 \quad \forall l \in \mathbb{N} \right\}, \quad X_+ = \left\{ z \in X : z_l > 0 \quad \forall l \in \mathbb{N} \right\}. \quad (16)$$

We cite some results of [10].

Proposition 1 (Ball, Carr, Penrose) *The space X is a Banach space, and it is the dual space of*

$${}^*X = \left\{ z = (z_l)_{l \in \mathbb{N}} : l^{-1} z_l \xrightarrow{l \rightarrow \infty} 0 \right\}. \quad (17)$$

Moreover, let $Z = (m \mapsto z^{(m)})$ be any sequence in X , and let $z^{(\infty)}$ be some element of X . Then

- (1) Z converges to $z^{(\infty)}$ weak* in X if and only if
 - (a) the sequence $m \mapsto \|z^{(m)}\|_X$ is bounded, and
 - (b) $z_l^{(m)} \xrightarrow{m \rightarrow \infty} z_l^{(\infty)}$ for all $l \in \mathbb{N}$.
- (2) Z converges to $z^{(\infty)}$ strongly in X if and only if
 - (a) $z^{(m)} \xrightarrow{m \rightarrow \infty} z^{(\infty)}$ weak* in X , and
 - (b) $\|z^{(m)}\|_X \xrightarrow{m \rightarrow \infty} \|z^{(\infty)}\|_X$.

Remarks. (i) The flux J_l is always weak* continuous for $l \geq 1$. (ii) Assumption (A2) provides the weak* continuity of J_0 . (iii) Assumption (A1) implies that the sequence $l \mapsto |l^{-1} \ln q_l|$ is bounded, and the availability functional A from (8) is thus well defined on the whole cone X_{0+} . (iv) The cone X_{0+} is closed under both strong and weak* convergence, and with (1) we have $\|z\|_X = \varrho(z)$ for all $z \in X_{0+}$.

For later purposes we define weak* continuous functionals N_l , $l \geq 1$, by

$$N_l(z) := \sum_{n=l}^{\infty} z_n. \quad (18)$$

Clearly, this definition implies $N(z) = N_1(z)$ and $z_l = N_l(z) - N_{l+1}(z)$. Moreover, by means of formal transformations we find $\varrho(z) = \sum_{l=1}^{\infty} N_l(z)$ and

$$\frac{d}{dt} N_l(z(t)) = J_{l-1}(z(t)) \quad \text{for all } l \in \mathbb{N} \cup \{0\}. \quad (19)$$

The existence of solutions for the modified model can be proved similarly to the classical results in [10]: In the first step we consider a finite, m -dimensional approximate problem, which results from the infinite system by neglecting all droplets with more than m atoms. This gives rise to the following system of ordinary differential equations

$$\begin{aligned}\frac{d}{dt}z_l^{(m)}(t) &= J_{l-1}(z^{(m)}(t)) - J_l(z^{(m)}(t)), \quad l = 2, \dots, m-1, \\ \frac{d}{dt}z_m^{(m)}(t) &= J_{m-1}(z^{(m)}(t)), \\ \frac{d}{dt}z_1^{(m)}(t) &= -J_1(z^{(m)}(t)) - \sum_{l=1}^{m-1} J_l(z^{(m)}(t)),\end{aligned}\tag{20}$$

with initial condition

$$z_l^{(m)}(0) = \tilde{z}_l^{(m)}, \quad \tilde{z}_l^{(m)} = \tilde{z}_l \quad \text{for } l = 1, \dots, m.\tag{21}$$

In the second step we construct weak solutions of the infinite system (MBD1)–(MBD3) as weak* limits of solutions to (20)–(21).

Remarks. (i) The vector $z^{(m)}$ can be regarded as an element of X by setting $z_l^{(m)} \equiv 0$ for all $l > m$. (ii) The approximate system is again closed by (MBD3). (iii) The initial data $\tilde{z}^{(m)}$ of the approximate system converge for $m \rightarrow \infty$ strongly in X to \tilde{z} , the initial data of the infinite system.

Existence and uniqueness results for the finite dimensional IVP (20)–(21) can be established by means of standard techniques for ODEs.

Lemma 2 *For all $m \in \mathbb{N}$ there exists a smooth and nonnegative solution $z^{(m)} \in C^\infty([0, \infty); X)$ of the approximate IVP (20)–(21). Moreover, with*

$$N_l^{(m)} = N(z^{(m)}), \quad J_l^{(m)} = J_l(z^{(m)}), \quad A^{(m)} = A(z^{(m)}), \quad \varrho^{(m)} = \varrho(z^{(m)})\tag{22}$$

we find $\varrho^{(m)}(t) = \varrho^{(m)}(0)$ and

$$-\frac{d}{dt}A^{(m)}(t) \geq \frac{\text{const}}{(\varrho^{(m)})^2} \sum_{l=1}^{m-1} |J_l^{(m)}(t)|^2 \quad \text{with } \text{const} > 0,\tag{23}$$

for all $t \geq 0$, and $\frac{d}{dt}N_l^{(m)}(t) = J_{l-1}^{(m)}(t)$ for all $t \geq 0$ and all $l = 1, \dots, m$.

Proof. For brevity we prove only (23). With similar transformations as in (9) and exploiting (MBD3) we obtain

$$\begin{aligned} \frac{d}{dt}A(z) &= \sum_{l=1}^{m-1} \gamma_l \left(z_1^{(m)} z_l^{(m)} - N_1^{(m)} \frac{q_l}{q_{l+1}} z_{l+1}^{(m)} \right) \ln \left(\frac{q_l z_{l+1}^{(m)} N_1^{(m)}}{q_{l+1} z_l^{(m)} z_1^{(m)}} \right) \\ &= - \sum_{l=1}^{m-1} \gamma_l (d_l - c_l) (\ln c_l - \ln d_l) \end{aligned} \quad (24)$$

with $c_l = z_1^{(m)} z_l^{(m)}$ and $d_l = N_1^{(m)} z_{l+1}^{(m)} q_l / q_{l+1}$. From $\lim_{l \rightarrow \infty} q_l / q_{l+1} = R$ and

$$z_l^{(m)} \leq \varrho^{(m)} / l \quad \text{and} \quad z_1^{(m)} \leq N_1^{(m)} \leq \varrho^{(m)} \quad (25)$$

it follows that $c_l, d_l < \text{const} (\varrho^{(m)})^2 / l$, and hence

$$\begin{aligned} \gamma_l (c_l - d_l) (\ln c_l - \ln d_l) &\geq \frac{\gamma_l}{\max\{c_l, d_l\}} (c_l - d_l)^2 \\ &\geq \frac{l \gamma_l}{\text{const} (\varrho^{(m)})^2} (c_l - d_l)^2 \\ &\geq \frac{\text{const}}{(\varrho^{(m)})^2} \frac{l}{\gamma_l} |J_l^{(m)}|^2. \end{aligned} \quad (26)$$

Assumption (A2) implies $l/\gamma_l \geq \text{const} > 0$, and (23) follows from (26). \square

In order to pass to the limit $m \rightarrow \infty$ we need some uniform estimates for the solution of the approximate problem.

Lemma 3 *The following functions in Lemma 2 are uniformly, i.e. independently of m , bounded in $C([0, \infty))$.*

- (1) $z_l^{(m)}, N_l^{(m)}, J_l^{(m)}, \dot{z}_l^{(m)}$, and $\dot{N}_l^{(m)}$ for all $l \geq 1$,
- (2) $\ddot{z}_l^{(m)}, \ddot{N}_l^{(m)}, \dot{J}_l^{(m)}$ for all $l \geq 2$,
- (3) $J_0^{(m)}, \dot{J}_1^{(m)}$.

For brevity we omit the proof, which is carried out in [11]. Moreover, we can derive all assertions quite easily from the equations (20) and assumption (A2).

Theorem 4 *Let $z^{(m)}$ as in Lemma 2. Then there exists a subsequence $j \mapsto z^{(m_j)}$, and a function $z \in C(I; X)$, $I = [0, \infty)$, with the following properties.*

- (1) (a) *The convergences $z_l^{(m)} \xrightarrow{j \rightarrow \infty} z_l$ and $N_l^{(m)} \xrightarrow{j \rightarrow \infty} N_l(z)$ are strong in $C(I)$ for $l = 1$, and even strong in $C^1(I)$ for $l \geq 2$,*
- (b) *The convergence $J_l^{(m)} \xrightarrow{j \rightarrow \infty} J_l(z)$ is strong in $C(I)$ for $l = 0$, and even strong in $C^1(I)$ for $l \geq 1$.*

- (2) We have $z_l(t) \geq 0$ for all $l \geq 1$ and all $t \in I$,
(3) The limit z satisfies

$$\frac{d}{dt} z_l(t) = J_{l-1}(z(t)) - J_l(z(t)), \quad \frac{d}{dt} N_l(z(t)) = J_{l-1}(z(t)), \quad (27)$$

for all $l \geq 2$ and all $t \in [0, \infty)$, and for all $t_1, t_2 \in I$ we have

$$z_1(t_2) - z_1(t_1) = \int_{t_1}^{t_2} J_0(z(t)) dt. \quad (28)$$

- (4) The availability A decreases according to

$$A(z(t_1)) - A(z(t_2)) \geq \frac{\text{const}}{(\varrho_0)^2} \int_{t_1}^{t_2} \sum_{l=1}^{\infty} |J_l(z(t))|^2 dt \geq 0. \quad (29)$$

Theorem 5 *The total mass of z from Theorem 4 is conserved, i.e. $\varrho(z(t)) = \varrho(z(0)) = \varrho_0$ for all finite $t \geq 0$.*

Remarks.

- (1) Because of (28) the limit z is a *weak* solution of (MBD1)–(MBD3).
- (2) In Section 4 it turns out to be useful that (27) holds in a strong sense for large l .
- (3) Inequality (29) follows from (23) and the Lemma of Fatou. All other assertions of Theorem 4 are consequences of the uniform bounds in Lemma 3 and the Arzelá-Ascoli Theorem, see [11]. Moreover, we obtain uniform continuity with respect to time for several functions including z_l , $N_l(z)$, $J_l(z)$ for $l \geq 1$.
- (4) The proof of Theorem 5 is not so obvious and needs some careful estimates for the mass contained in the tail of the solution. However, since one can use similar methods as in [10] we skip the proof and refer to [11].

Finally we give a brief summary of the uniqueness results in [11]. To establish uniqueness for the infinite system (MBD1)–(MBD3) it is convenient to pass to new variables $\zeta = (\zeta_l)_{l \in \mathbb{N}}$ with

$$\zeta_l := N_l(z) = \sum_{n=l}^{\infty} z_n. \quad (30)$$

Note that $z_l = \zeta_l - \zeta_{l+1}$, $N(z) = \zeta_1$, and $\varrho(z) = \sum_{l=1}^{\infty} \zeta_l$. The change of variables transforms (MBD1)–(MBD2) into

$$\frac{d}{dt} \zeta_l(t) = J_{l-1}(\zeta(t)) \quad \text{for } l \geq 1, \quad (31)$$

$$J_0(\zeta) = - \sum_{l=1}^{\infty} J_l(\zeta) \quad (32)$$

$$J_l(\zeta) = \gamma_l \left((\zeta_1 - \zeta_2) (\zeta_l - \zeta_{l+1}) - \zeta_1 \frac{q_l}{q_{l+1}} (\zeta_{l+1} - \zeta_{l+2}) \right) \quad \text{for } l \geq 1. \quad (33)$$

Note that Theorems 4 and 5 yield the global existence of weak solutions for (31)–(33). The reformulation of the original system now provides uniqueness results in form of Gronwall type estimates.

Theorem 6 *Let $\zeta^{(1)}$ and $\zeta^{(2)}$ be two weak solutions of (31)–(33), and set $\tilde{\zeta} = \zeta^{(2)} - \zeta^{(1)}$. Then there exists a time dependent constant $C(t)$ such that*

$$\|\tilde{\zeta}(t)\|_{\ell^1(\mathbb{N})} \leq \|\tilde{\zeta}(0)\|_{\ell^1(\mathbb{N})} + C(t) \int_0^t \|\tilde{\zeta}(s)\|_{\ell^1(\mathbb{N})} ds.$$

A similar result for the classical Becker–Döring equations is derived in [12], and the basic estimates therein can easily be adapted for proving Theorem 6. This is done in [11].

3 Equilibrium states

An equilibrium state of the Becker–Döring system is a state $\bar{z} \in X_+$, such that all fluxes J_l vanish in \bar{z} . Clearly, $0 \in X$ is always an equilibrium state. In this section we study equilibrium states with prescribed positive total mass $\varrho(\bar{z}) = \bar{\varrho}$. Here $\bar{\varrho} > 0$ is a given constant which remains fixed within this section.

For the analysis it is convenient to use the following variant \tilde{A} of the availability

$$\tilde{A}(z) = A(z) - \varrho(z) \ln R = \sum_{l=1}^{\infty} z_l \ln \left(\frac{z_l}{\tilde{q}_l N(z)} \right), \quad (34)$$

with $\tilde{q}_l = q_l R^l$ and R as in (A1), because \tilde{A} is weak* continuous on X_{0+} . To

prove this, we split \tilde{A} into three parts $\tilde{A} = \tilde{A}_1 + \tilde{A}_2 + \tilde{A}_3$, where

$$\tilde{A}_1(z) = -N(z) \ln(N(z)), \quad (35)$$

$$\tilde{A}_2(z) = \sum_{l=1}^{\infty} z_l \ln z_l, \quad (36)$$

$$\tilde{A}_3(z) = -\sum_{l=1}^{\infty} z_l \ln \tilde{q}_l. \quad (37)$$

The weak* continuity of \tilde{A}_1 is obvious, of \tilde{A}_2 it was proved in [10], and of \tilde{A}_3 it is a consequence of Proposition 1 and $\lim_{l \rightarrow \infty} l^{-1} \ln \tilde{q}_l = \ln 1 = 0$.

Next we derive a necessary condition for the existence of an equilibrium state \bar{z} with prescribed total mass $\varrho(\bar{z}) = \bar{\varrho} > 0$. We set $J_l(\bar{z}) = 0$ in (MBD3), and obtain

$$\bar{z}_{l+1} = \frac{\bar{z}_1}{\bar{N}} \frac{q_{l+1}}{q_l} \bar{z}_l = \frac{\bar{z}_1}{R \bar{N}} \frac{\tilde{q}_{l+1}}{\tilde{q}_l} \bar{z}_l, \quad (38)$$

where $\bar{N} = N(\bar{z})$. With $\tilde{q}_1 = R q_1 = R$ and the abbreviation $\bar{\mu} := \bar{z}_1 / (R \bar{N})$, $\bar{\mu} \in [0, 1/R]$, equation (38) yields

$$\bar{z}_l = \left(\frac{\bar{z}_1}{R \bar{N}} \right)^{l-1} \frac{\tilde{q}_l}{\tilde{q}_1} \bar{z}_1 = \bar{N} \tilde{q}_l \bar{\mu}^l = \bar{N} q_l (R \bar{\mu})^l \quad \text{for all } l \in \mathbb{N}. \quad (39)$$

Finally, the condition $\bar{N} = N(\bar{z})$ and the constraint $\varrho(\bar{z}) = \bar{\varrho}$ require

$$\bar{N} \sum_{l=1}^{\infty} \tilde{q}_l \bar{\mu}^l = \bar{N} \quad \text{as well as} \quad \bar{N} \sum_{l=1}^{\infty} l \tilde{q}_l \bar{\mu}^l = \bar{\varrho},$$

which imply

$$\tilde{f}(\bar{\mu}) = 1 \quad \text{and} \quad \bar{N} = \frac{\bar{\varrho}}{\tilde{g}(\bar{\mu})} \quad \text{with} \quad \tilde{f}(\mu) = \sum_{l=1}^{\infty} \tilde{q}_l \mu^l, \quad \tilde{g}(\mu) = \sum_{l=1}^{\infty} \tilde{q}_l l \mu^l. \quad (40)$$

Note that both power series in (40) have the same radius of convergence $\tilde{R} = 1$. The function \tilde{f} is continuous and strictly increasing on $[0, 1]$, and satisfies $\tilde{f}(\mu) \geq \tilde{q}_1 \mu = \mu R$. Consequently, the parameter $\bar{\mu}$ exists in the interval $[0, \min\{1, 1/R\}]$ if and only if $\tilde{f}(1) = \lim_{\mu \rightarrow 1} \tilde{f}(\mu) \geq 1$. Moreover, in order to guarantee $\varrho(\bar{z}) = \bar{\varrho} > 0$ we must have $\bar{N} > 0$, or equivalently, $\tilde{g}(\bar{\mu}) < \infty$. Since $\tilde{f}(1) > 1$ implies $\bar{\mu} < 1$ and therefore $\tilde{g}(\bar{\mu}) < \infty$, we end up with the following condition (EQ) for the existence of an equilibrium state

$$\tilde{f}(1) > 1, \quad \text{or} \quad \tilde{f}(1) = 1 \quad \text{and} \quad \tilde{g}(1) < \infty. \quad (\text{EQ})$$

Its negation reads

$$\tilde{f}(1) < 1, \quad \text{or} \quad \tilde{f}(1) = 1 \quad \text{and} \quad \tilde{g}(1) = \infty. \quad (\text{NEQ})$$

Theorem 7 For any $\bar{\varrho} > 0$ there exists an equilibrium state \bar{z} with $\varrho(\bar{z}) = \bar{\varrho}$ if and only if (EQ) is satisfied. Moreover, if (EQ) is satisfied then

(a) there exists a unique value $\bar{\mu} \in (0, 1]$ such that

$$\tilde{f}(\bar{\mu}) = 1, \quad (41)$$

(b) $\bar{z} \in X_+$ is given as in (39)–(40), i.e.

$$\bar{z}_l = \bar{N} \tilde{q}_l \bar{\mu}^l, \quad \bar{N} = N(\bar{z}) = \bar{\varrho} / \tilde{g}(\bar{\mu}), \quad (42)$$

(c) we have $\tilde{A}(\bar{z}) = \bar{\varrho} \ln \bar{\mu} \leq 0$.

For the two examples from Section 1 the equilibrium condition (EQ) reads as follows. *Example 1.* Equation (12) implies $R = \exp(-\delta) = \tilde{q}_1 < 1$ and

$$\begin{aligned} \tilde{f}(1) &= \exp(-\delta) + \sum_{l=2}^{\infty} \exp(-\gamma l^{2/3}) \\ &\leq \exp(-\delta) + \frac{1}{\gamma^{3/2}} \int_1^{\infty} \exp(-s^{2/3}) ds. \end{aligned} \quad (43)$$

In particular, for large values⁴ of both δ and γ there is no equilibrium state \bar{z} . *Example 2.* From (13) we deduce $R = \exp(+\beta) > 1$, $\tilde{q}_l = 1$ for large l , and $\tilde{f}(1) = \infty$, so that there always exists the equilibrium state (41)–(42) with $\bar{\mu} < 1/R < 1$.

Let $\partial_z \tilde{A}(z)$ and $\partial_z \varrho(z)$ denote the Gateaux differentials of A and ϱ in z , respectively, which are well defined for strictly positive $z \in X_+$. By means of basic calculus we derive from (42) that

$$\partial_z \tilde{A}(\bar{z}) = \left(\ln \frac{\bar{z}_l}{\tilde{q}_l \bar{N}} \right)_{l \in \mathbb{N}} = \left(\ln \bar{\mu}^l \right)_{l \in \mathbb{N}}, \quad \partial_z \varrho(\bar{z}) = (l)_{l \in \mathbb{N}}, \quad (44)$$

and conclude that (42) is equivalent to

$$\partial_z \tilde{A}(\bar{z}) = (\ln \bar{\mu}) \partial_z \varrho(\bar{z}). \quad (45)$$

However, since the functional \tilde{A} is not convex, it is not obvious that (45) defines a minimizer of \tilde{A} under the constraint of prescribed mass. For this reason we study the optimization problem

$$\tilde{A}_{\min} = \inf \left\{ \tilde{A}(z) : z \in X_{0+}, \varrho(z) = \bar{\varrho} \right\} \quad (\text{OPT})$$

in more detail. Our main results are formulated in the next two theorems.

⁴ See [3] for physically relevant values.

Theorem 8 For (EQ) the infimum \tilde{A}_{\min} in (OPT) is attained. Moreover, a minimizer is given by equations (41)–(42).

Theorem 9 For (NEQ) we have $\tilde{A}_{\min} = 0$ in (OPT), but there is no minimizer.

3.1 Proof of Theorem 8

Lemma 10 For $z \in X_{0+}$ and any $\mu \in (0, 1)$ we have

$$\tilde{A}(z) \geq \varrho(z) \ln \mu - N(z) \ln \left(\tilde{f}(\mu) \right). \quad (46)$$

Proof. It is sufficient to consider $z \neq 0$, so that $N(z) > 0$. At first we rewrite $T := \tilde{A}(z) - \varrho(z) \ln \mu$ as follows

$$\begin{aligned} T &= \tilde{A}(z) - \sum_{l=1}^{\infty} z_l \ln(\mu^l) = \sum_{l=1}^{\infty} z_l \ln \left(\frac{z_l}{\tilde{q}_l N(z) \mu^l} \right) \\ &= N(z) \sum_{l=1}^{\infty} (\tilde{q}_l \mu^l) \left(\frac{z_l}{\tilde{q}_l N(z) \mu^l} \right) \ln \left(\frac{z_l}{\tilde{q}_l N(z) \mu^l} \right) \\ &= N(z) \left(\sum_{l=1}^{\infty} p_l \right) \left(\sum_{l=1}^{\infty} p_l h(y_l) \right) / \left(\sum_{l=1}^{\infty} p_l \right), \end{aligned} \quad (47)$$

where $h(y) = y \ln y$, $p_l = \tilde{q}_l \mu^l$, and $y_l = z_l / (\tilde{q}_l N(z) \mu^l)$. Note that

$$\sum_{l=1}^{\infty} p_l = \tilde{f}(\mu) < \infty, \quad (48)$$

and $p_l > 0$ for all l . Since the function h is convex, Jensen's inequality yields

$$\begin{aligned} T &\geq N(z) \left(\sum_{l=1}^{\infty} p_l \right) h \left(\left(\sum_{l=1}^{\infty} p_l y_l \right) / \left(\sum_{l=1}^{\infty} p_l \right) \right) \\ &= N(z) \left(\sum_{l=1}^{\infty} p_l \right) h \left(\left(\sum_{l=1}^{\infty} z_l / N(z) \right) / \left(\sum_{l=1}^{\infty} p_l \right) \right) \\ &= N(z) \tilde{f}(\mu) h(1/\tilde{f}(\mu)) = -N(z) \ln \left(\tilde{f}(\mu) \right), \end{aligned} \quad (49)$$

and the proof is complete. \square

Corollary 11 Suppose (EQ), and let $z \in X_{0+}$ with $\varrho(z) = \bar{\varrho}$. Then,

$$\tilde{A}(z) \geq \bar{\varrho} \ln(\bar{\mu}) = \tilde{A}(\bar{z}), \quad (50)$$

where $\bar{\mu}$ and \bar{z} as in Theorem 7. In particular, Theorem 8 is proved.

Proof. Set $\mu = \bar{\mu}$ in Lemma 10, and compare with (c) in Theorem 7. \square

3.2 Proof of Theorem 9

In this section we consider the case (NEQ), i.e. we assume either $\tilde{f}(1) < 1$ or $\tilde{f}(1) = 1$ and $\tilde{g}(1) = \infty$, and we prove that now the optimization problem (OPT) has no minimizer. Recall that $\lim_{l \rightarrow \infty} \tilde{q}_l^{1/l} = 1$, and note that $\tilde{f}(1) \leq 1$ implies $\tilde{q}_l \leq 1$ for all $l \in \mathbb{N}$, as well as $\lim_{l \rightarrow \infty} \tilde{q}_l = 0$.

Our strategy is to construct certain perturbations of \tilde{q} , such that we can rely on the result of the previous section. For this reason we set

$$\Pi = \left\{ p = (p_l)_{l \in \mathbb{N}} : p_l > 0 \forall l \in \mathbb{N}, \quad \limsup_{l \rightarrow \infty} |l^{-1} \ln p_l| < \infty \right\}, \quad (51)$$

and define a functional \mathcal{A} on $X_{0+} \times \Pi$ by

$$\mathcal{A}(z, p) = \sum_{l=1}^{\infty} z_l \ln \frac{z_l}{p_l N(z)} = -N(z) \ln(N(z)) + \sum_{l=1}^{\infty} z_l \ln \frac{z_l}{p_l}, \quad (52)$$

so that $\tilde{A}(z) = \mathcal{A}(z, \tilde{q})$ and $A(z) = \mathcal{A}(z, q)$. Note that $\mathcal{A}(z, p)$ is well defined for all $(z, p) \in X_{0+} \times \Pi$. Moreover, if $\lim_{l \rightarrow \infty} p_l^{1/l} = 1$ the functional \mathcal{A} is weak* continuous with respect to z .

Definition (52) implies

$$\mathcal{A}(z, p^{(2)}) = \mathcal{A}(z, p^{(1)}) + \sum_{l=1}^{\infty} z_l \ln \frac{p_l^{(1)}}{p_l^{(2)}} \quad (53)$$

where $p^{(1)}, p^{(2)}$ are two arbitrary elements of Π . Furthermore, $-\mathcal{A}$ preserves the order in Π , i.e.

$$\mathcal{A}(z, p^{(2)}) \geq \mathcal{A}(z, p^{(1)}) \quad \text{for } p^{(2)} \leq p^{(1)}. \quad (54)$$

Now we approximate \tilde{q} by a sequence $(m \mapsto q^{(m)}) \subset \Pi$, where $q^{(m)}$ is defined by

$$q_l^{(m)} = \max \{ \tilde{q}_l, \pi_m \}, \quad \pi_m = \sup_{l > m} \tilde{q}_l. \quad (55)$$

Note that $\lim_{m \rightarrow \infty} \pi_m = 0$ and that $0 < \pi_m \leq 1$ for all $m \in \mathbb{N}$. If m is large the sequence $q^{(m)}$ is a good approximation of \tilde{q} , because both series differ only for large l . In particular,

$$l_m := \min \{ l : \tilde{q}_l \neq q_l^{(m)} \} = \min \{ l : \tilde{q}_l < \pi_m \} \xrightarrow{m \rightarrow \infty} \infty. \quad (56)$$

If \tilde{q} is a decreasing sequence, as for instance in the first example from Section 1, equation (56) reduces to

$$q_l^{(m)} = \tilde{q}_l \quad \text{for } l \leq m, \quad q_l^{(m)} = \tilde{q}_{m+1} \quad \text{for } l > m. \quad (57)$$

For any $m \in \mathbb{N}$ there exists a unique minimizer of $\mathcal{A}(\cdot, q^{(m)})$, because we find

$$\sum_{l=1}^{\infty} q_l^{(m)} = \infty > 1 \quad \text{and} \quad \lim_{l \rightarrow \infty} (q_l^{(m)})^{1/l} = \lim_{l \rightarrow \infty} (\pi_m)^{1/l} = 1, \quad (58)$$

and thus there exist variants of Theorems 7 and 8 with $q^{(m)}$ instead of \tilde{q} . This provides the existence of

$$A_{\min}^{(m)} := \min \left\{ \mathcal{A}(z, q^{(m)}) : \varrho(z) = \bar{\varrho} \right\}, \quad (59)$$

as well as the identity

$$A_{\min}^{(m)} = \bar{\varrho} \ln \mu_m = \mathcal{A}(z^{(m)}, q^{(m)}), \quad (60)$$

where $\mu_m \in (0, 1)$ and $z^{(m)} \in X_+$ satisfy

$$\sum_{l=1}^{\infty} q_l^{(m)} \mu_m^l = 1, \quad z_l^{(m)} = N_m q_l^{(m)} \mu_m^l, \quad (61)$$

where $N_m = \bar{\varrho} / (\sum_{l=1}^{\infty} q_l^{(m)} l \mu_m^l)$. Recall that $\varrho(z^{(m)}) = \bar{\varrho}$ for all m .

Definition (55) implies

$$\tilde{q} \leq \dots \leq q^{m+1} \leq q^m \leq \dots \leq q^1 \leq 1. \quad (62)$$

This chain and (54) give

$$\mathcal{A}(z, \tilde{q}) \geq \dots \geq \mathcal{A}(z, q^{m+1}) \geq \mathcal{A}(z, q^m) \geq \dots \geq \mathcal{A}(z, q^1) \quad \forall z \in X_{0+}, \quad (63)$$

and hence

$$\tilde{A}_{\min} \geq \dots \geq A_{\min}^{(m+1)} \geq A_{\min}^{(m)} \geq \dots \geq A_{\min}^{(1)}, \quad (64)$$

where $\tilde{A}_{\min} = \inf \left\{ \mathcal{A}(z, \tilde{q}) : \varrho(z) = \bar{\varrho} \right\}$.

Lemma 12

- (a) For any $z \in X_{0+}$ and $m \rightarrow \infty$ we have $\mathcal{A}(z, q^{(m)}) \uparrow \mathcal{A}(z, \tilde{q})$.
- (b) The sequence $m \mapsto z^{(m)}$ from (61) is a minimizing sequence for \tilde{A} , and $A_{\min}^{(m)} \uparrow \tilde{A}_{\min}$ for $m \rightarrow \infty$,

(c) $\tilde{A}_{\min} = 0$.

Proof. Using (53) and $q_l^{(m)} \leq 1$ we find

$$\begin{aligned} \mathcal{A}(z, \tilde{q}) - \mathcal{A}(z, q^{(m)}) &= \sum_{l=1}^{\infty} z_l \ln \frac{q_l^{(m)}}{\tilde{q}_l} \\ &= \sum_{l: \tilde{q}_l \neq q_l^{(m)}} z_l \ln \frac{q_l^{(m)}}{\tilde{q}_l} \leq \sum_{l: \tilde{q}_l \neq q_l^{(m)}} z_l \ln \frac{1}{\tilde{q}_l}, \end{aligned} \quad (65)$$

and Hölder's inequality gives

$$\begin{aligned} \mathcal{A}(z, \tilde{q}) - \mathcal{A}(z, q^{(m)}) &\leq \left(\sup_{l: \tilde{q}_l \neq q_l^{(m)}} |l^{-1} \ln \tilde{q}_l| \right) \left(\sum_{l: \tilde{q}_l \neq q_l^{(m)}} z_l l \right) \\ &\leq \left(\sup_{l: \tilde{q}_l \neq q_l^{(m)}} |\ln \tilde{q}_l^{1/l}| \right) \left(\sum_{l=1}^{\infty} z_l l \right) \leq \bar{\varrho} \sup_{l \geq l_m} |\ln \tilde{q}_l^{1/l}|, \end{aligned} \quad (66)$$

where l_m is defined in (56). Combining (66) and (63) yields

$$\mathcal{A}(z, q^{(m)}) \leq \mathcal{A}(z, \tilde{q}) \leq \mathcal{A}(z, q^{(m)}) + \bar{\varrho} \eta_m, \quad (67)$$

where η_m abbreviates $\eta_m = \sup_{l \geq l_m} |l^{-1} \ln \tilde{q}_l|$. The limit $\lim_{l \rightarrow \infty} \tilde{q}_l^{1/l} = 1$ implies $\lim_{l \rightarrow \infty} |l^{-1} \ln \tilde{q}_l| = 0$, and thanks to (56) we find $\lim_{m \rightarrow \infty} \eta_m = 0$. Since (63) provides the monotonicity as well as the convergence of the sequence $m \mapsto \mathcal{A}(z, q^{(m)})$ we can pass to the limit $m \rightarrow \infty$ in (67), and obtain assertion (a). Evaluating (67) for $z = z^{(m)}$ gives

$$A_{\min}^{(m)} \leq \mathcal{A}(z^{(m)}, \tilde{q}) \leq A_{\min}^{(m)} + \bar{\varrho} \eta_m. \quad (68)$$

Moreover, (64) implies

$$A_{\min}^{(m)} \leq \tilde{A}_{\min} \leq \mathcal{A}(z^{(m)}, \tilde{q}) \leq A_{\min}^{(m)} + \bar{\varrho} \eta_m. \quad (69)$$

Assertion (b) now follows from passing to the limit $m \rightarrow \infty$ in (69). Finally we prove assertion (c). From (62) and (61)₁ we derive

$$\mu_1 \leq \dots \leq \mu_m \leq \mu_{m+1} \leq \dots \leq 1. \quad (70)$$

Thus there exists $\tilde{\mu} := \lim_{m \rightarrow \infty} \mu_m \leq 1$. Suppose for contradiction that $\tilde{\mu} < 1$. Then,

$$\left(m \mapsto (\mu_m^l)_{l \in \mathbb{N}} \right) \xrightarrow{m \rightarrow \infty} (\tilde{\mu}^l)_{l \in \mathbb{N}} \quad \text{in } \ell^1(\mathbb{N}). \quad (71)$$

Since $q^{(m)}$ converges for $m \rightarrow \infty$ to \tilde{q} in $\ell^\infty(\mathbb{N})$, we can conclude

$$1 = \sum_{l=1}^{\infty} q_l^{(m)} \mu_m^l \xrightarrow{m \rightarrow \infty} \sum_{l=1}^{\infty} \tilde{q}_l \tilde{\mu}^l < \sum_{l=1}^{\infty} \tilde{q}_l \leq 1. \quad (72)$$

This contradiction shows $\tilde{\mu} = 1$. Therefore

$$\tilde{A}_{\min} = \lim_{m \rightarrow \infty} A_{\min}^{(m)} = \bar{\varrho} \lim_{m \rightarrow \infty} \ln \mu_m = 0, \quad (73)$$

which was claimed. \square

Corollary 13 *Since (EQ) is violated, there is no minimizer in (OPT). In particular, Theorem 9 is proved.*

Proof. By contradiction. Suppose there is a state $z \in X_{0+}$ with $\varrho(z) = \bar{\varrho} > 0$ and $\tilde{A}(z) = 0$. Then $z \neq 0$ and hence $N(z) > 0$. According to Lemma 10 we can estimate

$$0 = \tilde{A}(z) \geq \varrho(z) \ln \mu - N(z) \ln \tilde{f}(\mu) \quad \text{for all } \mu \in (0, 1). \quad (74)$$

At first suppose $\tilde{f}(1) < 1$, and let $\mu \rightarrow 1$. Then (74) yields a contradiction, namely $0 = \tilde{A}(z) \geq -N(z) \ln \tilde{f}(1) > 0$. Now suppose $\tilde{f}(1) = 1$ and $\tilde{g}(1) = \infty$. Then, (74) implies

$$\varrho(z) \ln \mu \leq N(z) \ln \tilde{f}(\mu) \leq \varrho(z) \ln \tilde{f}(\mu) \quad (75)$$

and hence $\mu \leq \tilde{f}(\mu)$ for all $\mu \in (0, 1)$. Moreover, from $\mu \tilde{f}'(\mu) = \tilde{g}(\mu)$ we conclude $\lim_{\mu \rightarrow 1} \tilde{f}'(\mu) = \infty$. Therefore, for μ_1 and μ_2 with $\mu_1, \mu_2 \lesssim 1$ we find

$$\tilde{f}(\mu_2) - \tilde{f}(\mu_1) = \int_{\mu_1}^{\mu_2} \tilde{f}'(\mu) d\mu \geq 2(\mu_2 - \mu_1). \quad (76)$$

With $\mu_2 \rightarrow 1$ it follows

$$\tilde{f}(\mu_1) \leq \tilde{f}(1) - 2(1 - \mu_1) = 2\mu_1 - 1 < \mu_1, \quad (77)$$

which is the desired contradiction. \square

Corollary 14 *Let $m \mapsto z^{(m)}$ be an arbitrary sequence of minimizers for problem (OPT). Then, $z^{(m)} \xrightarrow{m \rightarrow \infty} 0$ weak* in X_{0+} .*

Proof. The sequence is bounded and thus weak* compact. Let $j \mapsto z^{(m_j)}$ be a subsequence, such that $z^{(m_j)} \rightarrow z^{(\infty)}$ weak* in X_{0+} for $j \rightarrow \infty$. The weak* continuity of \tilde{A} implies $\tilde{A}(z^{(\infty)}) = 0$. Suppose that $z^{(\infty)} \neq 0$, i.e.

$\varrho_\infty := \varrho(z^{(\infty)}) > 0$, and let $\tilde{z} = \bar{\varrho} z^{(\infty)} / \varrho_\infty$. Since $\tilde{A}(\tilde{z}) = \bar{\varrho} \tilde{A}(z^{(\infty)}) / \varrho_\infty = 0$, Corollary 13 yields a contradiction. We conclude $z^{(\infty)} = 0$, which shows that 0 is the unique accumulation point of the sequence. This implies the claimed convergence. \square

4 The limit $t \rightarrow \infty$.

In this section we study the longtime behavior of the solution $t \mapsto z(t)$ from Section 2. At first we show that any final limit is an equilibrium state, and then we investigate whether this state is unique, and whether the mass remains conserved in the limit $t \rightarrow \infty$. Recall that $\varrho(z(t)) = \varrho(z(0)) = \varrho_0$ holds for all finite times $t \geq 0$.

4.1 Auxiliary result

Lemma 15 *For all $l \geq 0$ and $t \rightarrow \infty$ we have $J_l(z(t)) \rightarrow 0$.*

Proof. Suppose for contradiction that there exist some $\varepsilon > 0$, an index $l_0 \geq 1$, and a sequence $m \mapsto t_m$ with $t_m \rightarrow \infty$ for $m \rightarrow \infty$, such that

$$|J_{l_0}(z(t_m))| \geq 2\varepsilon \quad \text{for all } m \in \mathbb{N}. \quad (78)$$

The uniform continuity of $t \mapsto J_{l_0}(z(t))$, see the remarks for Theorems 4 and 5, imply the existence of $\tau > 0$ with

$$|J_{l_0}(z(t))| \geq \varepsilon \quad \text{for all } m \in \mathbb{N} \text{ and } t \in (t_m, t_m + \tau). \quad (79)$$

By extracting a subsequence, still denoted by $m \mapsto t_m$, we can achieve that $t_m + \tau \leq t_{m+1}$ for all $m \in \mathbb{N}$. Estimate (29) now implies

$$A(z(t_m)) - A(z(t_{m+1})) \geq \frac{\text{const}}{\varrho_0^2} \int_{t_m}^{t_{m+1}} |J_{l_0}(z(t))|^2 dt \geq \tau\varepsilon, \quad (80)$$

and hence $A(z(t_m)) \rightarrow -\infty$ for $m \rightarrow \infty$, which contradicts either Theorem 8 or Theorem 9. This proves the assertion for all $l \neq 0$. Now suppose $l_0 = 0$ in (78). Without loss of generality we can assume that there exists $z^{(\infty)} \in X_{0+}$ such that

$$z^{(m)} \xrightarrow{m \rightarrow \infty} z^{(\infty)} \quad \text{weak}^* \text{ in } X, \quad (81)$$

which implies $J_l(z^{(m)}) \xrightarrow{m \rightarrow \infty} J_l(z^{(\infty)})$ for all $l \geq 0$, because all functionals J_l are weak* continuous, see the remarks for Proposition 1. In the first part of this proof we have shown that $J_l(z^{(\infty)}) = 0$ for all $l \geq 1$. Finally, we find

$$J_0(z^{(\infty)}) = - \sum_{l=1}^{\infty} J_l(z^{(\infty)}) = 0, \quad (82)$$

which is a contradiction for (78). \square

Let X_w be the space X equipped with with the weak* topology, and let ω denote the ω -limit set of $Z = \{z(t) : t \geq 0\}$ in X_w , i.e.

$$\omega = \left\{ z \in X : z = \text{weak}^* \text{-} \lim_{m \rightarrow \infty} z(t_m) \text{ for some } m \mapsto t_m \text{ with } \lim_{m \rightarrow \infty} t_m = \infty \right\}.$$

Corollary 16 *Z is relatively compact in X_w , and ω contains at least one equilibrium state $z^{(\infty)} \in X_{0+}$ with*

$$\varrho_{\infty} := \varrho(z^{(\infty)}) \in [0, \varrho_0] \quad \text{and} \quad \tilde{A}_{\infty} := \tilde{A}(z^{(\infty)}) = \lim_{t \rightarrow \infty} \tilde{A}(z(t)). \quad (83)$$

Moreover, if $z^{(\infty)}$ is unique, then

$$z(t) \xrightarrow{t \rightarrow \infty} z^{(\infty)} \quad \text{weak}^* \text{ in } X, \quad (84)$$

and this convergence is strong in X if and only if $\varrho_{\infty} = \varrho_0$.

Proof. Note that $\lim_{t \rightarrow \infty} \tilde{A}(t)$ exists, because the function $t \mapsto \tilde{A}(t)$ is decreasing and bounded. Moreover, since the total mass is conserved, the set Z is relatively compact in X_w , and therefore ω contains some $z^{(\infty)} \in X$, which clearly satisfies $z^{(\infty)} \in X_{0+}$. Lemma 15 shows that $z^{(\infty)}$ is in fact an equilibrium state, and (83) comes from the weak* continuity of \tilde{A} . The remaining assertions follow from elementary topological principles and Proposition 1. \square

Next we prove a sufficient condition for the mass conservation in the limit $t \rightarrow \infty$.

Theorem 17 *Suppose that there exist $R' \in [0, R)$ and a time $t_0 \geq 0$ such that*

$$\lambda(t) := z_1(t)/N(t) \leq R' \quad (85)$$

holds for all $t \geq t_0$. Then the mass must be conserved for $t \rightarrow \infty$, i.e. $\varrho(z^{(\infty)}) = \varrho_0$ for all $z^{(\infty)}$ from Corollary 16.

Note that (85) is equivalent to $\Gamma_l^V(t) \geq \kappa \Gamma_l^C(t)$, $\kappa = R'/R$, so that the assumption of Theorem (17) implies that for large cluster and large times the fragmentation process dominates coagulation.

4.2 Main Results

Before we prove Theorem 17 we discuss its consequences. To this end we consider several cases which are gathered in the following table.

Case	Conditions	Limit $t \rightarrow \infty$	Convergence
<i>NEQ</i> :	(NEQ)	$z^{(\infty)} = 0$	weak*
<i>EQ-1</i> :	(EQ), $\tilde{f}(1) = 1$	OPEN	
<i>EQ-2</i> :	(EQ), $\tilde{f}(1) > 1$, $R > 1$	$z^{(\infty)} = \bar{z}(\varrho_0)$	strong
<i>EQ-3</i> :	(EQ), $\tilde{f}(1) > 1$, $R \leq 1$		
<i>EQ-3a</i> :	..., $\tilde{A}_\infty = 0$,	$z^{(\infty)} = 0$	weak*
<i>EQ-3b</i> :	..., $\tilde{A}_\infty < 0$,	$z^{(\infty)} = \bar{z}(\varrho_0)$	strong

In the case *NEQ* the solution converges weak* to 0, because there is no equilibrium state with positive mass at all. This case is actually the most interesting one, because here all mass is contained in larger and larger clusters when time increases. The same phenomenon occurs within the standard model if ϱ_0 exceeds the critical value ϱ_S , and in this case the long-time dynamics of the large clusters is governed by the Lifshitz–Slyozov–Wagner (LSW) equation, see [13] for a formal derivation, and [14,12,9,15] for rigorous results. We expect to find an analogue for the LSW equation, now describing the long time evolution for case *NEQ* of the modified model. However, this problem is addressed in a forthcoming paper.

Let us continue with *EQ-2*. As we will see below, the condition $R > 1$ implies the conservation of mass for $t \rightarrow \infty$ without further assumption, i.e. $z^{(\infty)}$ is the unique equilibrium state with mass ϱ_0 , and all claimed results follow immediately from Corollary 16.

Next we consider *EQ-3*. Theorem 7 provides a family of equilibrium states \bar{z} , which are parameterized by the total mass ϱ , or alternatively, by the availability \tilde{A} . Corollary 16 states $z^{(\infty)} = \bar{z}(\tilde{A}_\infty)$, i.e. $z^{(\infty)}$ is the unique equilibrium state with availability \tilde{A}_∞ , so that the uniqueness of \tilde{A}_∞ implies the uniqueness of $z^{(\infty)}$. Now suppose *EQ-3b*. From $\tilde{A}_\infty < 0$ it follows that $\varrho_\infty \neq 0$, and we will see that this already gives $\varrho_\infty = \varrho_0$, cf. Corollary 18 below. However, in subcase *EQ-3a* we have $\tilde{A}_\infty = 0$, i.e. $z^{(\infty)} = \bar{z}(0) = 0$, and thus we con-

clude that this subcase is very similar to *NEQ*. Note that the initial condition $\tilde{A}(z(0)) < 0$ surely implies *EQ-3b*. However, for $\tilde{A}(z(0)) > 0$ it may depend on the distribution of the initial mass whether the long time behavior is governed by *EQ-3a* or *EQ-3b*.

Finally we discuss *EQ-1*. Again there exists a family of possible equilibrium states, see Theorem 7. However, all these equilibrium states have the same availability, because $\bar{\mu} = 1$ implies $\tilde{A}(\bar{z}) = \bar{\varrho} \ln \bar{\mu} = 0$, and it remains open to establish uniqueness.

Now we have formulated all results concerning the limit $t \rightarrow \infty$. In the remaining part we prove Theorem 17 as well as the following result.

Corollary 18 *In the cases *EQ-2* and *EQ-3b* we have $\varrho(z^{(\infty)}) = \varrho_0$.*

Proof. For Case *EQ-2* we set $t_0 = 0$ and choose $R' \in (1, R)$. Then, Theorem 17 provides the conservation of mass, because $\lambda(t)$ takes values in $[0, 1]$. Now suppose Case *EQ-3b*, and recall that $\tilde{A}(z^{(\infty)}) < 0$ implies $z^{(\infty)} \neq 0$, and hence $N(z^{(\infty)}) > 0$. Since $z(t) \rightarrow z^{(\infty)}$ weak* in X for $t \rightarrow \infty$, we find

$$\lambda(t) = \frac{z_1(t)}{N(z(t))} \xrightarrow{t \rightarrow \infty} \frac{z_1^{(\infty)}}{N(z^{(\infty)})} = \tilde{q}_1 \bar{\mu} =: \bar{\lambda}, \quad (86)$$

and $\tilde{f}(1) > 1$ gives $\bar{\mu} < 1$ and $\bar{\lambda} < R$. Consequently, the assumptions of Theorem 17 are satisfied for $R' \in (\bar{\lambda}, R)$ and t_0 sufficiently large. \square

The proof of Theorem 17 consists of several non-trivial steps, which we present in the following two subsections. Before we go into details, we shall briefly describe the main ideas. At first we recall the quantities ζ_l from Section 2

$$\zeta_l = N_l(z) = \sum_{n=l}^{\infty} z_n, \quad (87)$$

such that $\zeta_1 = N(z)$ and $\varrho(z) = \sum_{l=1}^{\infty} \zeta_l$. In what follows we will identify certain sequences $\sigma = (\sigma_l)_{l \in \mathbb{N}}$ for which

$$H_\sigma(t) := \max \left\{ \frac{\varrho_0}{\sigma_{l_0}}, \sup_{l \geq l_0+1} \frac{\zeta_l(t)}{\sigma_l} \right\} \quad (88)$$

decreases with time for $t \geq t_0$ and for l_0 sufficiently large. Moreover, some of these sequences σ are elements of $\ell^1(\mathbb{N})$. Consequently, for all $\varepsilon > 0$ there exists an index l_1 , such that $\sum_{l=l_1}^{\infty} \zeta_l(t) \leq H_\sigma(t_0) \sum_{l=l_1}^{\infty} \sigma_l \leq \varepsilon$ holds true for all $t \geq t_0$, and this uniform estimate implies the conservation of mass for $t \rightarrow \infty$.

The approach described above is inspired by Ball and Carr [16], which use a similar idea to prove conservation of mass within the standard model. Another application of the method from [16] is given in [17].

4.3 More auxiliary results

Let $l_0 \in \mathbb{N}$ be given, and let $\eta = (\eta_l)_{l \in \mathbb{N}}$ be any strictly positive sequence with

$$0 < \eta_0 := \sup_{l \geq l_0} \eta_l < 1. \quad (89)$$

Depending on l_0 and η we define a set $S = S_{\eta, l_0}$ by

$$S := \left\{ \sigma = (\sigma_l)_{l \in \mathbb{N}} : \begin{array}{l} (i) \ \sigma_l \geq \sigma_{l+1} \geq 0 \text{ for all } l \in \mathbb{N}, \\ (ii) \ (\sigma_l - \sigma_{l+1}) \geq \eta_l (\sigma_{l-1} - \sigma_l) \text{ for all } l \geq l_0 \end{array} \right\}. \quad (90)$$

Moreover, let $S_+ := S \setminus \{0\}$ and note that (90) provides $\sigma_l > 0$ for all $\sigma \in S_+$ and all $l \in \mathbb{N}$.

Lemma 19 (Ball, Carr) *The set S is closed under (i) addition, (ii) multiplication with positive constants, (iii) pointwise convergence of sequences, and (iv) taking pointwise infima in arbitrary subsets.*

Proof. (i) – (iii) are obvious. To prove (iv), let I be an arbitrary index set and $I \ni i \mapsto \sigma^{(i)}$ be any family in S . We set $\sigma_l = \inf_{i \in I} \sigma_l^{(i)}$ for all $l \in \mathbb{N}$. By construction,

$$\sigma_l^{(i)} \geq \sigma_l \quad \text{for all } i \in I, l \in \mathbb{N}, \quad (91)$$

$$\sigma_l^{(i)} \geq \sigma_{l+1}^{(i)} \geq 0 \quad \text{for all } i \in I, l \in \mathbb{N}, \quad (92)$$

$$\left(\sigma_l^{(i)} - \sigma_{l+1}^{(i)} \right) \geq \eta_l \left(\sigma_{l-1}^{(i)} - \sigma_l^{(i)} \right) \quad \text{for all } i \in I, l \geq l_0. \quad (93)$$

(91) and (92) imply $\sigma_l^{(i)} \geq \sigma_{l+1}^{(i)} \geq \sigma_{l+1} \geq 0$ and hence $\sigma_l \geq \sigma_{l+1} \geq 0$. (91) and (93) yield

$$(1 + \eta_l) \sigma_l^{(i)} \geq \sigma_{l+1}^{(i)} + \eta_l \sigma_{l-1}^{(i)} \geq \sigma_{l+1} + \eta_l \sigma_{l-1},$$

and thus $(1 + \eta_l) \sigma_l \geq \sigma_{l+1} + \eta_l \sigma_{l-1}$, as required. \square

We say, a sequence $\sigma \in S$ is a *S-majorant* of a sequence $\xi \in \ell^\infty(\mathbb{N})$, if $\sigma_l \geq \xi_l$ holds for all $l \geq l_0$.

Corollary 20 *There exists an operator $\widehat{\cdot} : \ell^\infty(\mathbb{N}) \rightarrow S$, mapping ξ to $\widehat{\xi}$, such that for given ξ the image $\widehat{\xi}$ is the minimal S -majorant of ξ . This reads*

$$\widehat{\xi} = \inf \left\{ \sigma \in S : \sigma_l \geq \xi_l \quad \text{for all } l \geq l_0 \right\}. \quad (94)$$

Proof. The set in which we take the infimum in (94) is not empty, because it contains at least sufficiently large constants. The remaining assertions are due to Lemma 19. \square

Lemma 21 *Let $m \in \mathbb{N}$, and let $\delta^{(m)}$ be the Dirac distribution in m , i.e. $\delta_m^{(m)} = 1$ and $\delta_l^{(m)} = 0$ for all $l \neq m$. Then, $\widehat{\delta^{(m)}} \in S_+$ satisfies $\widehat{\delta^{(m)}}_l = 1$ for all $l \leq m$. Moreover, we have*

$$\|\widehat{\delta^{(m)}}\|_{\ell^\infty(\mathbb{N})} = 1 \quad \text{as well as} \quad \|\widehat{\delta^{(m)}}\|_{\ell^1(\mathbb{N})} \leq m + \frac{\eta_0}{1 - \eta_0}, \quad (95)$$

where η_0 is given in (89).

Proof. At first we observe that $\widehat{\delta^{(m)}}$ must decrease, so that $\widehat{\delta^{(m)}}_l \geq 1$ for all $l \leq m$. Next we define a S -majorant σ of $\delta^{(m)}$ by $\sigma_l = 1$ for $l \leq m$, and $\sigma_l = \eta_0^{l-m}$ for $l \geq m$. Clearly, we have $\sigma \in S$ as well as

$$\|\sigma\|_{\ell^\infty(\mathbb{N})} = 1 \quad \text{and} \quad \|\sigma\|_{\ell^1(\mathbb{N})} = m + \frac{\eta_0}{1 - \eta_0}. \quad (96)$$

Since $\widehat{\delta^{(m)}}$ is the minimal S -majorant of $\delta^{(m)}$, we conclude $\sigma \geq \widehat{\delta^{(m)}}$, and this implies all claimed results. \square

Lemma 22 (Ball, Carr) *Let $\xi \in \ell^\infty(\mathbb{N})$ be arbitrary. Then $\lim_{l \rightarrow \infty} \xi_l = 0$ implies $\lim_{l \rightarrow \infty} \widehat{\xi}_l = 0$.*

Proof. Since $\widehat{\xi}$ is a nonnegative and increasing sequence there exists the limit

$$2\varepsilon := \lim_{l \rightarrow \infty} \widehat{\xi}_l = \inf_{l \in \mathbb{N}} \widehat{\xi}_l. \quad (97)$$

Suppose for contradiction that $\varepsilon > 0$. Then there exists $l_1 \in \mathbb{N}$ such that for all $l > l_1$ we have $\xi_l \leq \varepsilon < 2\varepsilon \leq \widehat{\xi}_l$. Due to Lemma 21 there is at least one strictly positive sequence $\sigma \in S$ with $\lim_{l \rightarrow \infty} \sigma_l = 0$. For any $b > 0$ the sequence $\varepsilon + b\sigma$ is contained in S , and for sufficiently large b it is a S -majorant of ξ , but not of $\widehat{\xi}$. This is the desired contradiction. \square

In the next section we need the following stronger version of Lemma 22.

Lemma 23 Any nonnegative and decreasing sequences $\xi \in \ell^\infty(\mathbb{N})$ satisfies

$$\sum_{l=1}^{\infty} \xi_l < \infty \iff \sum_{l=1}^{\infty} \widehat{\xi}_l < \infty. \quad (98)$$

Proof. (\Leftarrow) is obvious. To prove (\Rightarrow) let ξ be nonnegative and decreasing with $\xi \in \ell^1(\mathbb{N})$. We define a sequence $m \mapsto \sigma^{(m)}$ of sequences as follows

$$\sigma^{(m)} := \left(\sum_{k=1}^{m-1} (\xi_k - \xi_{k+1}) \widehat{\delta^{(k)}} \right) + \xi_m \widehat{\delta^{(m)}}, \quad (99)$$

where $\widehat{\delta^{(k)}}$ as in Lemma 21. Clearly, for all m we have $\sigma^{(m)} \in S$ and

$$\begin{aligned} \sigma_l^{(m)} &= \left(\sum_{k=1}^{m-1} (\xi_k - \xi_{k+1}) \widehat{\delta^{(k)}}_l \right) + \xi_m \widehat{\delta^{(m)}}_l \\ &\geq \left(\sum_{k=l}^{m-1} (\xi_k - \xi_{k+1}) \widehat{\delta^{(k)}}_l \right) + \xi_m \widehat{\delta^{(m)}}_l \\ &= \left(\sum_{k=l}^{m-1} \xi_k - \xi_{k+1} \right) + \xi_m = \xi_l \end{aligned} \quad (100)$$

with $l = 1, \dots, m-1$. Moreover, from $\widehat{\delta^{(m+1)}} \geq \delta^{(m)}$ it follows $\widehat{\delta^{(m+1)}} \geq \widehat{\delta^{(m)}}$ and hence

$$\sigma^{(m+1)} = \sigma^{(m)} - \xi_{m+1} \widehat{\delta^{(m)}} + \xi_{m+1} \widehat{\delta^{(m+1)}} \geq \sigma^{(m)}. \quad (101)$$

In particular, there exists the pointwise limit $\sigma^{(\infty)} := \lim_{m \rightarrow \infty} \sigma^{(m)}$. Lemma 19 gives $\sigma^{(\infty)} \in S$, and with $\xi \leq \sigma^{(\infty)}$ we find $\widehat{\xi} \leq \sigma^{(\infty)}$. Finally, Lemma 21 provides the following uniform estimate

$$\begin{aligned} \|\sigma^{(m)}\|_{\ell^1(\mathbb{N})} &\leq \xi_m \|\widehat{\delta^{(m)}}\|_{\ell^1(\mathbb{N})} + \sum_{k=1}^{m-1} (\xi_k - \xi_{k+1}) \|\widehat{\delta^{(k)}}\|_{\ell^1(\mathbb{N})} \\ &\leq \xi_m \left(m + \frac{\eta_0}{1 - \eta_0} \right) + \sum_{k=1}^{m-1} (\xi_k - \xi_{k+1}) \left(k + \frac{\eta_0}{1 - \eta_0} \right) \\ &= \frac{\xi_1 \eta_0}{1 - \eta_0} + \xi_m m + \sum_{k=1}^{m-1} (\xi_k - \xi_{k+1}) k \\ &= \frac{\xi_1 \eta_0}{1 - \eta_0} + \xi_m m + \left(-(m-1) \xi_m + \sum_{k=1}^{m-1} \xi_k \right) \\ &= \frac{\xi_1 \eta_0}{1 - \eta_0} + \sum_{k=1}^m \xi_k \leq \frac{\xi_1 \eta_0}{1 - \eta_0} + \sum_{k=1}^{\infty} \xi_k, \end{aligned} \quad (102)$$

and the Lemma of Fatou yields $\sigma^{(\infty)} \in \ell^1(\mathbb{N})$, which implies $\widehat{\xi} \in \ell^1(\mathbb{N})$. \square

We mention that the equivalence (98) may fail if ξ is not decreasing.

4.4 Proof of Theorem 17

Within this subsection we always assume that the assumptions of Theorem 17 are satisfied, i.e. we have $\lambda(t) \leq R'$ for some $R' < R$ and for all $t \geq t_0$.

Lemma 24 *There exists $\mu_0 < 1$ and an index $l_0 \in \mathbb{N}$ such that*

$$\frac{d}{dt} \zeta_l(t) \leq N(z(t)) \gamma_{l-1} \frac{q_{l-1}}{q_l} \left(\mu_0 (\zeta_{l-1}(t) - \zeta_l(t)) - (\zeta_l(t) - \zeta_{l+1}(t)) \right) \quad (103)$$

is satisfied for all $l \geq l_0$ and all $t \geq t_0$.

Proof. Recall from (A1) that $R = \lim_{l \rightarrow \infty} q_l/q_{l+1}$. We choose μ_0 and l_0 such that $\lambda(t) \leq \mu_0 q_l/q_{l+1}$ holds for all $t \geq t_0$ and all $l \geq l_0$. This implies

$$\begin{aligned} J_l(z(t)) &= \gamma_l N(z(t)) \frac{q_l}{q_{l+1}} \left(\frac{q_{l+1}}{q_l} \lambda(t) z_l(t) - z_{l+1}(t) \right) \\ &\leq \gamma_l N(z(t)) \frac{q_l}{q_{l+1}} (\mu_0 z_l - z_{l+1}(t)) \end{aligned} \quad (104)$$

for all $t \geq t_0$ and $l \geq l_0 - 1$. From Theorem 4 we read off

$$\frac{d}{dt} \zeta_l(t) = J_{l-1}(z(t)), \quad (105)$$

and with $z_l(t) = \zeta_l(t) - \zeta_{l+1}(t)$ we obtain (103). \square

Now we can make use of the auxiliary results from the previous subsection. For this reason we fix μ_0 , t_0 and l_0 as in Lemma 24, and we define the set S as in Equation (90), where the sequence η is assumed to be constant with value μ_0 .

Our next aim is to prove that for all $\sigma \in S_+$ the quantity $H_\sigma(t)$ from (88) decreases with time t . For any time t with $t \geq t_0$ we define a set $S(t) \subseteq S_+$ and a sequence $\hat{\sigma}(t) \in S$ by

$$S(t) := \left\{ \sigma \in S_+ : \sigma_l \geq \zeta_l(t') \quad \forall l \geq l_0 \text{ and } \forall t' \in [t, \infty) \right\}, \quad (106)$$

$$\hat{\sigma}(t) := \inf S(t). \quad (107)$$

Since $S(t)$ contains at least the constant ϱ_0 , $\hat{\sigma}(t) \in S$ is well defined and satisfies $\hat{\sigma}(t) \leq \varrho_0$. We mention that $t_1 < t_2$ implies $S(t_1) \subseteq S(t_2)$ and hence $\hat{\sigma}(t_1) \geq \hat{\sigma}(t_2)$.

For technical reasons we introduce some discrete counterparts of $S(t)$ and $\hat{\sigma}(t)$. For fixed $m \in \mathbb{N}$ with $m \geq l_0$ we define

$$\begin{aligned} S^{(m)}(t) &:= \left\{ \sigma \in S_+ : \sigma_l \geq \zeta_l(t') \ \forall l \text{ with } l_0 \leq l \leq m+1 \text{ and } \forall t' \in [t, \infty) \right\}, \\ \hat{\sigma}^{(m)}(t) &:= \inf S^{(m)}(t). \end{aligned} \quad (108)$$

Obviously, $\hat{\sigma}^{(m)}(t) \leq \hat{\sigma}(t)$, and again we find $\hat{\sigma}^{(m)}(t_1) \geq \hat{\sigma}^{(m)}(t_2)$ for all $t_1 < t_2$. The sequence $m \mapsto \hat{\sigma}^{(m)}(t)$ is increasing and bounded for all t , because $m_1 < m_2$ gives $\hat{\sigma}(t) \geq \hat{\sigma}^{(m_2)}(t) \geq \hat{\sigma}^{(m_1)}(t)$. This implies the existence of the pointwise limit $\lim_{m \rightarrow \infty} \hat{\sigma}^{(m)}(t) \leq \hat{\sigma}(t)$. Moreover, since this limit is an S -majorant of $\zeta(t')$ for all $t' \geq t$, it follows

$$\hat{\sigma}^{(m)}(t) \xrightarrow{m \rightarrow \infty} \hat{\sigma}(t) \quad \text{pointwise in } \ell^\infty(\mathbb{N}) \text{ for all } t \geq t_0. \quad (109)$$

Remark 25 *There exists $C \in \mathbb{R}$ such that $\limsup_{l \rightarrow \infty} \hat{\sigma}_l(t) l \leq C$ holds for all $t \geq t_0$.*

Proof. Let $l_1 \geq l_0$, and define a sequence σ by $\sigma_l = \varrho_0$ for $l \leq l_1$ and $\sigma_l = \varrho_0/l$ for $l > l_1$. For all $l > l_1$ we have

$$\frac{\sigma_l - \sigma_{l+1}}{\sigma_{l-1} - \sigma_l} = \frac{\frac{1}{l} - \frac{1}{l+1}}{\frac{1}{l-1} - \frac{1}{l}} = \frac{(l-1)}{(l+1)} \geq \frac{(l_1-1)}{(l_1+1)} \quad (110)$$

Next we choose l_1 sufficiently large such that $(l_1 - 1)/(l_1 + 1) > \mu_0$, and we find $\sigma \in S_+$. Let $t' \geq t$ be arbitrary. By definition we have

$$\zeta_l(t') = \sum_{n=l}^{\infty} z_n(t) \leq \frac{1}{l} \sum_{n=l}^{\infty} n z_n(t) \leq \frac{\varrho_0}{l}, \quad (111)$$

i.e. σ is an S -majorant of $\zeta(t')$. Hence $\hat{\sigma}(t) \leq \sigma$. Finally, $C := \varrho_0 l_1$ completes the proof. \square

Remark 26 *For fixed $m \geq l_0$, all $t \geq t_0$ and arbitrary $\sigma \in S_+$ let*

$$H_\sigma^{(m)}(t) := \max \left\{ \frac{\hat{\sigma}_{l_0}^{(m)}(t)}{\sigma_{l_0}}, \frac{\hat{\sigma}_{m+1}^{(m)}(t)}{\sigma_{m+1}}, \max_{l=l_0+1, \dots, m} \frac{\zeta_l(t)}{\sigma_l} \right\}. \quad (112)$$

Then,

$$\sigma \in S^{(m)}(t) \quad \iff \quad H_\sigma^{(m)}(t') \leq 1 \quad \forall t' \in [t, \infty) \quad (113)$$

is satisfied for all $\sigma \in S_+$.

Proof. Within this proof let t' always be arbitrary in $[t, \infty)$. We start with (\Leftarrow). From $H_\sigma^{(m)}(t') \leq 1$ it follows

$$\sigma_l \geq \zeta_l(t') \quad \forall l = l_0 + 1, \dots, m, \quad (114)$$

and

$$\sigma_{l_0} \geq \hat{\sigma}_{l_0}^{(m)}(t'), \quad \sigma_{m+1} \geq \hat{\sigma}_{m+1}^{(m)}(t'). \quad (115)$$

Since $\hat{\sigma}_{l_0}^{(m)}(t') \geq \zeta_{l_0}(t')$ and $\hat{\sigma}_{m+1}^{(m)}(t') \geq \zeta_{m+1}(t')$ holds by construction, and since t' was arbitrary, we find $\sigma \in S^{(m)}(t)$. Next we prove (\Rightarrow). $\sigma \in S^{(m)}(t)$ gives $\sigma \in S^{(m)}(t')$, and thus $\sigma \geq \hat{\sigma}^{(m)}(t')$. This implies (114) as well as (115), and we conclude $H_\sigma^{(m)}(t') \leq 1$. \square

Lemma 27 *Let $m \geq l_0$ and $\sigma \in S_+$ be given. Then, the function $t \mapsto H_\sigma^{(m)}(t)$ from (112) is decreasing. In particular, any $\sigma \in S_+$ satisfies*

$$\hat{\sigma}^{(m)}(t) \leq H_\sigma^{(m)}(t) \sigma. \quad (116)$$

Proof. Note that the function $t \mapsto H_\sigma^{(m)}(t)$ is well defined and continuous for all $t \geq t_0$, and let $t_1 \geq t_0$ be fixed. We prove by contradiction that

$$H_\sigma^{(m)}(t) < H_\sigma^{(m)}(t_1) + \varepsilon \quad (117)$$

holds for all $\varepsilon > 0$ and all $t \geq t_1$. Let $\varepsilon > 0$ be fixed and suppose

$$H_\sigma^{(m)}(t_2) = H_\sigma^{(m)}(t_1) + \varepsilon =: H_\varepsilon^{(m)}. \quad (118)$$

for some $t_2 > t_1$ with $H_\sigma^{(m)}(t) < H_\varepsilon^{(m)}$ for all t with $t_1 \leq t < t_2$. We find

$$H_\varepsilon^{(m)} = \max_{l \geq l_0+1, \dots, m} \frac{\zeta_l(t_2)}{\sigma_l}, \quad (119)$$

because the functions $t \mapsto \hat{\sigma}_{l_0}^{(m)}(t)$ and $t \mapsto \hat{\sigma}_{l_0}^{(m+1)}(t)$ are decreasing. Thus there exists $l_1 \in \{l_0 + 1, \dots, m\}$ such that

$$\zeta_{l_1}(t_2) = H_\varepsilon^{(m)} \sigma_{l_1}. \quad (120)$$

Moreover, Definition (112) guarantees that

$$\zeta_l(t) \leq H^{(m)}(t) \sigma_l \leq H_\varepsilon^{(m)} \sigma_l \quad (121)$$

for all $l = l_0, \dots, m + 1$ and all $t \in [t_1, t_2]$. According to (103) we find

$$\begin{aligned} \frac{d}{dt} \zeta_l(t) &\leq N(z(t)) \gamma_l \frac{q_l}{q_{l+1}} \left(\mu_0 \zeta_{l-1}(t) - (1 + \mu_0) \zeta_l(t) + \zeta_{l+1}(t) \right) \\ &\leq N(z(t)) \gamma_l \frac{q_l}{q_{l+1}} \left(H_\varepsilon^{(m)} \left(\mu_0 \sigma_{l-1} + \sigma_{l+1} \right) - (1 + \mu_0) \zeta_l(t) \right) \\ &\leq \varrho_0 \gamma_l \frac{q_l}{q_{l+1}} (1 + \mu_0) \left(H_\varepsilon^{(m)} \sigma_l - \zeta_l(t) \right), \end{aligned} \quad (122)$$

where the last estimate is due to $\mu_0 \sigma_{l-1} + \sigma_{l+1} \leq (1 + \mu_0) \sigma_l$ which follows from the definition of S , see (90). We apply Gronwall's Lemma for $t \in [t_1, t_2]$, and obtain

$$\left(H_\varepsilon^{(m)} \sigma_l - \zeta_l(t_2) \right) \geq \exp \left(-c_l (t_2 - t_1) \right) \left(H_\varepsilon^{(m)} \sigma_l - \zeta_l(t_1) \right) > 0, \quad (123)$$

where $c_l > 0$ can be read off from (122). The estimate (123) with $l = l_1$ is a contradiction for (120). Thus we have proved (117), and the limit $\varepsilon \rightarrow 0$ yields the claimed monotonicity result. Finally, let $\sigma \in S_+$ be fixed and $t \geq t_1$ be arbitrary. We find

$$\zeta_l(t) \leq H_\sigma^{(m)}(t) \sigma \leq H_\sigma^{(m)}(t_1) \sigma \quad \text{for all } l = l_0, \dots, m + 1. \quad (124)$$

In particular, $H_\sigma^{(m)}(t_1) \sigma \in S^{(m)}(t_1)$, and it follows $\hat{\sigma}^{(m)}(t_1) \leq H_\sigma^{(m)}(t_1) \sigma$, which was claimed in (116). \square

Lemma 28 *For all $t \geq t_0$ we have $\hat{\sigma}(t) = \widehat{\eta(t)}$ where*

$$\eta(t) := \max \left\{ \zeta(t), \hat{\sigma}_{l_0}(t) \delta^{(l_0)} \right\}, \quad (125)$$

$\delta^{(l_0)}$ is the Dirac distribution in l_0 , and $\widehat{\eta(t)}$ is the minimal S -majorant of $\eta(t)$.

Proof. Let $m \geq l_0$ be arbitrary. Remark 26 and Lemma 27 provide

$$\begin{aligned} S^{(m)}(t) &= \left\{ \sigma \in S_+ : H^{(m)}(t) \leq 1 \right\} \\ &= \left\{ \sigma \in S_+ : \begin{array}{l} \sigma_{l_0} \geq \hat{\sigma}_{l_0}^{(m)}(t) \\ \sigma_l \geq \zeta_l(t) \quad \forall l = l_0 + 1, \dots, m \\ \sigma_{m+1} \geq \hat{\sigma}_{m+1}^{(m)}(t) \end{array} \right\}. \end{aligned} \quad (126)$$

Let $\eta^{(m)}(t) \in S_+$ be defined by

$$\eta^{(m)}(t) := \hat{\sigma}_{m+1}^{(m)}(t) + \widehat{\eta(t)}, \quad (127)$$

with $\eta(t)$ as in (125). As simple calculation shows $H_{\eta^{(m)}(t)}^{(m)}(t) \leq 1$, and from (116) it follows that

$$\hat{\sigma}^{(m)}(t) \leq H_{\eta^{(m)}(t)}^{(m)}(t) \eta^{(m)}(t) \leq \widehat{\eta}(t) + \hat{\sigma}_{m+1}(t). \quad (128)$$

According to Remark 25 and (109) the limit $m \rightarrow \infty$ provides

$$\hat{\sigma}(t) = \lim_{m \rightarrow \infty} \hat{\sigma}^{(m)}(t) \leq \widehat{\eta}(t). \quad (129)$$

Moreover, by construction we have $\hat{\sigma}_l(t) \geq \zeta_l(t)$ and $\hat{\sigma}_l(t) \geq \hat{\sigma}_{l_0} \delta_l^{(l_0)}$ for all $l \geq l_0$, which shows that $\hat{\sigma}(t)$ is S -majorant for $\eta(t)$. Therefore, $\hat{\sigma}(t) \geq \widehat{\eta}(t)$. \square

Corollary 29 *For all $t \geq t_0$ and all $\sigma \in S_+$ let $H_\sigma(t)$ be given as in (88), i.e.*

$$H_\sigma(t) := \max \left\{ \frac{\varrho_0}{\sigma_{l_0}}, \sup_{l \geq l_0+1} \frac{\zeta_l(t)}{\sigma_l} \right\}, \quad (130)$$

where $H_\sigma(t)$ may be infinite. Then, for all $\sigma \in S_+$ the function $t \mapsto H_\sigma(t)$ is decreasing. In particular, any $\sigma \in S_+$ satisfies

$$\hat{\sigma}(t) \leq H_\sigma(t) \sigma. \quad (131)$$

Proof. Let $\sigma \in S_+$ be fixed, let $t_1 \geq t_0$ with $H := H_\sigma(t_1) < \infty$, and let $t_2 \geq t_1$. With $\tilde{\sigma} = H\sigma$ we find $\tilde{\sigma}_{l_0} \geq \varrho_0 \geq \hat{\sigma}_{l_0}(t_1) \geq \zeta_{l_0}(t_1)$ and $\tilde{\sigma}_l \geq \zeta_l(t_1)$ for all $l > l_0$. In particular, $\tilde{\sigma}$ is an S -majorant of $\eta(t_1)$, and we conclude $\tilde{\sigma} \geq \widehat{\eta}(t_1)$. Lemma 28 yields $\tilde{\sigma} \geq \hat{\sigma}(t_1) \geq \hat{\sigma}(t_2)$, and it follows that $\tilde{\sigma}$ is an S -majorant of $\zeta(t_2)$. This implies $\tilde{\sigma}_l \geq \zeta_l(t_2)$ for all $l > l_0$, and hence

$$1 \geq H_{\tilde{\sigma}}(t_2) = \frac{1}{H} H_\sigma(t_2), \quad (132)$$

which was claimed. Finally, (131) follows immediately. \square

Corollary 30 *Let $z^{(\infty)}$ be as in Corollary 16, and let $\varepsilon > 0$ be arbitrary. Then we have*

$$\varrho_0 \geq \varrho(z^{(\infty)}) \geq \varrho_0 - 2\varepsilon \quad (133)$$

In particular, Theorem 17 is proved.

Proof. Let $\varepsilon > 0$ be fixed, and let

$$\sigma := H \widehat{\zeta}(t_0), \quad H := H_{\widehat{\zeta}(t_0)}(t_0) = \frac{\varrho_0}{\widehat{\zeta}(t_0)_{l_0}}. \quad (134)$$

We have $H \in [1, \infty)$, and Lemma 23 provides $\sigma \in \ell^1(\mathbb{N})$. Therefore we can choose an index $l_1 \geq l_0$ such that $\sum_{l=l_1}^{\infty} \sigma_l \leq \varepsilon$. Moreover, according to Corollary 29 we have $\zeta_l(t) \leq \sigma_l$ for all $l \geq l_0$ and all $t \geq t_0$. Therefore, we find

$$\varrho_0 = \sum_{l=1}^{\infty} \zeta_l(t) \leq \varepsilon + \sum_{l=1}^{l_1} \zeta_l(t) = \varepsilon + \sum_{l=1}^{l_1} N_l(z(t)). \quad (135)$$

By construction, there exists a sequence $m \mapsto t_m$ with $t_m \rightarrow \infty$ such that $z(t_m) \rightarrow z^{(\infty)}$ weak* in X for $m \rightarrow \infty$. Using the weak* continuity of the functionals N_l we find

$$\sum_{l=1}^{l_1} N_l(z^{(\infty)}) \geq \varrho_0 - \varepsilon. \quad (136)$$

Finally, it is easy to prove that $\varrho(z^{(\infty)}) \leq \varrho_0$ implies $\varrho(z^{(\infty)}) = \sum_{l=1}^{\infty} N_l(z^{(\infty)})$, and (133) follows immediately. \square

Acknowledgements

The authors would like to thank Wolfgang Dreyer and Frank Duderstadt for several fruitful discussions and for their valuable comments and remarks.

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