J. Nonlinear Sci. (2003): pp. 115–155
DOI: 10.1007/s00332-002-0535-8

Journal of
Nonlinear
Science

On the Evolution of Large Clusters in the Becker-Döring Model

B. Niethammer
Institut für Angewandte Mathematik, Universität Bonn, Wegelerstrasse 6,
53115 Bonn, Germany
e-mail: Barbara.Niethammer@iam.uni-bonn.de

Received August 5, 2002; accepted October 2, 2002
Online publication December 13, 2002
Communicated by P. J. Holmes

Summary. We consider the Becker-Döring equations for large times. It is well-known [2] that if the total density of monomers exceeds a critical value, the excess density is contained in larger and larger clusters as time proceeds. We rigorously derive for general coefficients that the evolution of these large clusters is described by a nonlocal transport equation, which is for specific coefficients the classical coarsening model by Lifshitz, Slyozov, and Wagner (LSW). Our proof exploits the estimate of the energy and the energy dissipation rate given by the Lyapunov functional for the Becker-Döring equations. We also provide a detailed asymptotic expansion of the higher-order dynamics.

1. Introduction

1.1. The Becker-Döring Equations

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 by gaining or losing one particle respectively. In particular, the Becker-Döring equations can be used to investigate various aspects in the kinetics of phase transitions, such as nucleation, metastability, and coarsening.

In the following, clusters are described by their size l, the number of atoms in the cluster. We denote by $c_l(t)$ the concentration of l-clusters at time t, and we assume that the clusters are uniformly distributed in space, such that there is no dependence on the space variable.

The crucial assumption in the Becker-Döring theory is that an l-cluster can change its size only by gaining a free atom (coagulation) to form an $(l+1)$-cluster, or lose an atom (fragmentation) to form an $(l-1)$-cluster. The net rate of conversion of l-clusters into $(l+1)$-clusters is denoted by J_l, which is measured in units of clusters per unit time per unit volume. The rate of change of the density of l-clusters is thus given by

$$\frac{d}{dt}c_l(t) = J_{l-1}(t) - J_l(t), \quad \text{for } l \geq 2. \tag{1.1}$$

To describe the rate of change of the density of free atoms, in the following called monomers, a different equation is needed since monomers are involved in the rate of change of all clusters. In the classical Becker-Döring theory [3] the concentration of monomers was just assumed to be constant. However, for the case of density-conserved systems, one would like to have the total number density of atoms to be preserved, i.e.,

$$\rho := \sum_{l=1}^{\infty} l c_l(t) \equiv const., \quad \text{for all } t \geq 0. \tag{1.2}$$

This implies with (1.1) that

$$\frac{d}{dt}c_1(t) = -J_1 - \sum_{l=1}^{\infty} J_l. \tag{1.3}$$

This modified version of the Becker-Döring equations was introduced in [4], [26].

To complete the system of equations, a constitutive relation which gives J_l in terms of c_l is required. For that, one assumes that the number of times an l-cluster gains a monomer per unit time per unit volume is proportional to the density of l-clusters and the density of monomers. The number of times that an $(l+1)$-cluster breaks up per unit time per unit volume is, however, independent of c_1 but proportional to c_{l+1}. Thus, one finds

$$J_l(t) = a_l c_1(t) c_l(t) - b_{l+1} c_{l+1}(t), \tag{1.4}$$

with positive kinetic coefficients a_l, b_l which are assumed to be independent of time. We assume in the following that the coefficients a_l, b_l are given by

$$a_l = l^{\alpha}, \qquad \text{for some } 0 \leq \alpha < 1, \tag{1.5}$$

$$b_l = a_l\left(z_s + \frac{q}{l^{\gamma}}\right), \quad \text{where } z_s > 0,\ q > 0 \text{ and } 0 < \gamma < 1, \tag{1.6}$$

which are the same assumptions as those used in [23], except that we also allow for $\alpha = 0$. These coefficients typically arise in density-conserving phase transitions as e.g. the formation of liquid droplets in a supersaturated vapor or the phase segregation in a binary alloy after quenching. The Becker-Döring equations apply to the case of a nonuniform mixture, i.e., when the saturation density is small or respectively when one component of the alloy has small volume fraction.

In Appendix A.1 we review the heuristic derivation of typical coefficients, when the clusters are spheres and the growth of clusters either is dominated by diffusion of monomers between the clusters or is limited by the reaction rate at the boundary of the cluster, respectively. One obtains (1.5), (1.6) with

$\alpha = 1/3$,	$\gamma = 1/3$	(diffusion-controlled kinetics in 3-D)
$\alpha = 0$,	$\gamma = 1/2$	(diffusion-controlled kinetics in 2-D)
$\alpha = 2/3$,	$\gamma = 1/3$	(interface-reaction-limited kinetics in 3-D)
$\alpha = 1/2$,	$\gamma = 1/2$	(interface-reaction-limited kinetics in 2-D)

Within these applications, z_s is just the equilibrium monomer concentration at a flat phase interface and q is proportional to surface tension. We also refer to [25] for a derivation of the coefficients for diffusion-controlled kinetics from an Ising model with Kawasaki dynamics. The relationship between this microscopic model and the Becker-Döring equations is also further investigated in [27], [28].

Different exponents can arise, for example, if the clusters cannot be reasonably characterized as spheres, but rather have the shape of snowflakes or are thin needles or plates.

1.2. Existence, Uniqueness, and Convergence to Equilibrium

We now summarize briefly the main mathematical results which have been obtained for the Becker-Döring equations with coefficients satisfying (1.5), (1.6). For a more extensive overview of the subject, we also refer to the review article [30].

The existence of positive solutions of the Becker-Döring equations has been shown in the seminal mathematical paper [2] under quite general assumptions on the coefficients, which are also satisfied by (1.5), (1.6). Uniqueness was shown only for a smaller class of coefficients, but more recently the uniqueness result has been extended to a larger class of coefficients in [15], which also covers the coefficients in (1.5), (1.6).

We now turn to the simplest solutions of the Becker-Döring equations, which are equilibrium solutions. By (1.1) equilibrium solutions $(\bar{c}_l)_l$ are given by $J_l \equiv const.$ for all l, but then due to (1.3) it must hold that

$$J_l = 0, \quad \text{for } l \geq 1.$$

This implies

$$\bar{c}_l = Q_l z^l, \quad l \geq 1,$$

with a parameter $z > 0$, where Q_l are given by

$$Q_1 = 1, \qquad \frac{Q_{l+1}}{Q_l} = \frac{a_l}{b_{l+1}}, \quad \text{and thus} \quad Q_l = \frac{a_1 a_2 \cdots a_{l-1}}{b_2 b_3 \cdots b_l}. \tag{1.7}$$

Depending on the coefficients, the equilibrium density $\sum_{l=1}^{\infty} l Q_l z^l$ is bounded for z in a certain range.

With the assumptions (1.5), (1.6) we easily obtain for large l that

$$Q_l \cong \frac{C_0}{l^\alpha z_s^{l-1}} \exp\left\{-\frac{q}{(1-\gamma)z_s} l^{1-\gamma}(1 + O(l^{-\gamma}))\right\}. \tag{1.8}$$

Then the series

$$\sum_{l=1}^{\infty} l Q_l z^l$$

has the convergence radius $\lim_{l\to\infty} \frac{b_{l+1}}{a_l} = z_s$ and the series also converges for $z = z_s$. In the following we denote

$$\rho_s := \sum_{l=1}^{\infty} l Q_l z_s^l < \infty, \tag{1.9}$$

which can be interpreted as the density of saturated vapor. In the following we will denote by $c_l^s = Q_l z_s^l$, $l = 1, 2, \ldots$, the equilibrium configuration with density ρ_s.

Convergence of solutions to equilibrium was shown in [2] under some assumptions on coefficients and data, and was further generalized in [1], [29]. The proof is based on the fact that there is a Lyapunov functional available, given by

$$L(c(t)) := \sum_{l=1}^{\infty} c_l \left(\ln \left(\frac{c_l}{Q_l} \right) - 1 \right). \tag{1.10}$$

In fact, it holds that

$$\frac{d}{dt} L(c(t)) = - \sum_{l=1}^{\infty} J_l \ln \left(\frac{a_l c_1 c_l}{b_{l+1} c_{l+1}} \right) \le 0.$$

Since L is bounded below, it follows that $J_l \to 0$ as $t \to \infty$ such that $c_l \to Q_l z^l$ for some $z > 0$. The question remains: What is z, and what happens to density conservation (1.2) in the limit as $t \to \infty$? It is shown in [2], [1], [29] that if $\rho \le \rho_s$ then

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

where $\rho = \sum_{l=1}^{\infty} l Q_l z^l$. However, if $\rho > \rho_s$, we have

$$\lim_{t \to \infty} c_l(t) = Q_l z_s^l, \quad \text{for each } l \ge 1,$$

but the convergence is only weak and the density drops to ρ_s in the limit $t \to \infty$. The so-called excess density is contained in larger and larger clusters as times evolves. In phase transformations, these large clusters represent the stable nuclei of the new thermodynamic phase, e.g., the liquid droplets formed out of the supersaturated vapor.

1.3. Metastability

In case $\rho > \rho_s$, i.e., when a phase transformation occurs in the sense described above, it turns out that the Becker-Döring model can describe another important feature, the occurrence of metastable states.

Existence of metastable states in the Becker-Döring model has been established in [23]. More precisely, specific solutions of the Becker-Döring equations are constructed, for data with density $\rho > \rho_s$, which stay very long, that is, at least exponentially long in $1/(\rho - \rho_s)$, close to the data, before they converge to their corresponding equilibrium. The crucial idea in the analysis is the following (for more details see [23], [30]): One looks for so-called steady-state solutions, such that $J_l \equiv const. \neq 0$. These are not exact solutions of the equation, but turn out to persist for a very long time. The common value of J_l is called the nucleation rate and gives the rate per unit volume at which clusters gain new clusters in the steady state. In fact, it is shown that if $c_1 - z_s$ is small, then the nucleation rate is extremely small, such that large clusters form extremely slowly.

Thus, this analysis gives an example of initial data for which a metastable state occurs. In fact, numerical simulations [5] suggest that the system always goes through a metastable state: If one starts with data with $c_1(0) = \rho$, then the value of c_1 drops quickly to a value larger than z_s, where it remains extremely long before finally converging to z_s.

It is interesting to note that metastability also occurs in the classical Becker-Döring equations where c_1 is constant, which is established in [23], [13]. We also refer to [10], [11] for a study of corresponding truncated systems.

1.4. Dynamics of Large Clusters: Heuristics

We now investigate the governing dynamics of the large clusters which form the new thermodynamic phase, once any possible metastable state has broken down.

For the special case of diffusion-controlled growth in three dimensions, i.e., $\alpha = \gamma = \frac{1}{3}$ in (1.5), (1.6), it is argued formally in [24] that the evolution of these clusters is governed by the classical coarsening model by Lifshitz, Slyozov, and Wagner [16], [33], nowadays known as the classical LSW model.

We recall briefly the argument in [24] for this case (see also [30]). To consider large times, we introduce a new time scale $\tau = \varepsilon t$ for a small parameter $\varepsilon \to 0$, such that

$$\frac{d}{d\tau} c_l = \frac{1}{\varepsilon}(J_{l-1} - J_l). \tag{1.11}$$

We write

$$J_l = a_l((c_1 - z_s) - q)c_l - (b_{l+1}c_{l+1} - b_l c_l), \tag{1.12}$$

and choose as a cut between small and large clusters $l_0 = l_0(\varepsilon)$, which can be chosen as $l_0 = \ln\left(\frac{1}{\varepsilon}\right)$ for example. For $l \geq l_0$ one substitutes $\lambda = \varepsilon l$ and treats λ as a continuous variable. Furthermore one introduces the rescaled cluster densities and fluxes as

$$c_l \ = \ \varepsilon^2 \nu(\lambda, \tau), \tag{1.13}$$

$$J_l \ = \ \varepsilon^2 \upsilon(\lambda, \tau), \tag{1.14}$$

and the rescaled monomer density

$$c_1 - z_s = \varepsilon^{1/3} u(\tau). \tag{1.15}$$

This gives

$$\begin{aligned} \partial_\tau \nu + \partial_\lambda \upsilon \ &= \ o(1), \\ \upsilon \ &= \ (\lambda^{1/3} u(\tau) - q)\nu + o(1). \end{aligned}$$

Now one considers the small clusters. It is argued in [24] that one can expect from (1.14) that $J_{l_0} = O(\varepsilon^2)$ and then from (1.11) that $J_l = O(\varepsilon l_0)$ for $l \leq l_0$. Furthermore, by solving (1.1) in terms of J_l, one obtains with (1.15) that

$$\begin{aligned} c_l \ &= \ Q_l c_1^l \left(1 - \sum_{k=1}^{l-1} \frac{J_k}{a_k Q_k c_1^k} \right) \\ &= \ Q_l z_s^l (1 + O(l\varepsilon^{1/3}))(1 + O(\varepsilon l_0^2 e^{\frac{3q}{2z_s} l_0^{2/3}})), \end{aligned}$$

and hence

$$c_l = Q_l z_s^l (1 + o(1)). \tag{1.16}$$

Density conservation (1.2) now gives

$$\begin{aligned}\rho = \sum_{l=1}^{\infty} l c_l &= \sum_{l=1}^{l_0-1} l c_l + \sum_{l=l_0}^{\infty} l c_l \\ &= \sum_{l=1}^{l_0-1} l Q_l z_s^l + \sum_{l=l_0}^{\infty} l c_l + o(1) \\ &= \rho_s + \int \lambda \nu(\lambda, \tau)\, d\lambda + o(1).\end{aligned}$$

Hence, one finds to leading order that

$$\partial_\tau \nu + \partial_\lambda((\lambda^{1/3} u(\tau) - q)\nu) = 0, \tag{1.17}$$

$$\int \lambda \nu \, d\lambda = \rho - \rho_s, \tag{1.18}$$

which implies

$$u(\tau) = \frac{q \int \nu \, d\lambda}{\int \lambda^{1/3} \nu \, d\lambda}.$$

The system (1.17), (1.18) is just the classical LSW model for coarsening (cf. [16], [33]). We will explain the scenario described by this model in more detail in Section 1.5.4 below.

Let us now briefly point out how the above argument applies to the case of general coefficients as given in (1.5), (1.6). For that $1/\varepsilon$ will again be a measure for the large clusters, but for general coefficients the time scale is given by $\tau = \varepsilon^{1-\alpha+\gamma} t$ and the rescaled clusters and fluxes by

$$c_l = \varepsilon^2 \nu, \qquad J_l = \varepsilon^{2+\alpha-\gamma} \upsilon, \qquad c_1 - z_s = \varepsilon^\gamma u.$$

We obtain to leading order

$$\partial_\tau \nu + \partial_\lambda \left(\lambda^\alpha \left(u - \frac{q}{\lambda^\gamma} \right) \nu \right) = 0, \tag{1.19}$$

$$\int \lambda \nu \, d\lambda = \rho - \rho_s, \tag{1.20}$$

which gives

$$u(\tau) = \frac{q \int \lambda^{\alpha-\gamma} \nu \, d\lambda}{\int \lambda^\alpha \nu \, d\lambda}. \tag{1.21}$$

1.5. Aims and Results of the Paper

1.5.1. Preliminary Considerations. A main goal in this paper is to justify the arguments in [24], respectively Section 1.4, by a rigorous analysis for the class of coefficients given in (1.5), (1.6). Our analysis will in particular also give an interpretation of the so-far ad hoc chosen parameter ε, which is related to the energy of the system.

Let us explain the main ideas in a bit more detail. The key idea is to exploit the estimate given by the Lyapunov functional (1.10), in physical terms the free energy density. To that aim we redefine the energy

$$F(c) := \sum_{l=1}^{\infty} c_l \left(\ln\left(\frac{c_l}{Q_l z_s^l} \right) - 1 \right) + Q_l z_s^l. \tag{1.22}$$

Since

$$F(c) = L(c) - \ln z_s \sum_{l=1}^{\infty} l c_l + \sum_{l=1}^{\infty} Q_l z_s^l$$

and $\sum_{l=1}^{\infty} l c_l$ is preserved during the evolution, we find $\frac{d}{dt}F = \frac{d}{dt}L$. Indeed, without the constant term $\sum_{l=1}^{\infty} Q_l z_s^l$, this is just the specific functional which is called V_{z_s} in [2] and is found to be continuous under weak$*$ convergence in the space of positive sequences which satisfy $\sum_{l=1}^{\infty} l c_l < \infty$. Our definition ensures that $F(c) \geq 0$ and $F(c) = 0$ if and only if $(c_l) = (Q_l z_s^l) = (c_l^s)$, i.e., if c is the equilibrium cluster distribution for the critical density ρ_s.

With this definition and the results in [2], [1], [29], we know that $F(c(t)) \to 0$ as $t \to \infty$.

It is also instructive to write F in the following way:

$$F(c(t)) = \sum_{l=1}^{\infty} c_l \left(\ln\left(\frac{c_l}{c_l^s} \right) - 1 \right) + c_l^s = \sum_{l=1}^{\infty} c_l^s f\left(\frac{c_l - c_l^s}{c_l^s} \right), \tag{1.23}$$

with $f(z) = (1+z)\ln(1+z) - z$, which resembles the notion often used in the study of the Boltzmann equation, where F is usually called the relative entropy. We also notice that f behaves quadratically for bounded $\frac{c_l - c_l^s}{c_l^s}$, whereas the growth is only superlinear for large $\frac{c_l - c_l^s}{c_l^s}$, which is the case for the large clusters.

We now ask for the leading order term in the energy for large clusters. Using (1.8), we find

$$\begin{aligned} \sum_{l=l_0}^{\infty} c_l \left(\ln\left(\frac{c_l}{c_l^s} \right) - 1 \right) + c_l^s &= \sum_{l=l_0}^{\infty} c_l \ln\left(\frac{1}{Q_l z_s^l} \right) + \sum_{l=l_0}^{\infty} c_l (\ln c_l - 1) + c_l^s \\ &\approx \frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} c_l + O\left(\sum_{l=l_0}^{\infty} l^{1-2\gamma} c_l \right) \\ &\quad + \sum_{l=l_0}^{\infty} c_l (\ln c_l - 1) + c_l^s. \end{aligned}$$

It is easily seen that $\sum_{l=l_0}^{\infty} c_l(\ln c_l - 1) = o(\sum_{l=l_0}^{\infty} l^{1-\gamma} c_l)$ and the same holds for $\sum_{l=l_0}^{\infty} c_l^s$ if l_0 is just moderately large, since (c_l^s) is decreasing exponentially fast.

Thus, the leading order term in the energy is

$$\frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} c_l,$$

which in the applications given in Section 1.1 is just the surface energy density of the clusters. Recalling the scaling introduced in Section 1.4, we find

$$\frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} c_l \approx \varepsilon^{\gamma} \frac{q}{z_s(1-\gamma)} \int \lambda^{1-\gamma} \nu \, d\lambda.$$

Hence, a natural criterion for the system being in the last stage is that the energy scales like ε^{γ} if ε^{-1} is a measure for the large clusters. (To be precise, it should be the other way: If the energy is small, the measure for the large clusters is given by the appropriate power of the energy.)

1.5.2. The Main Result. In the following we consider the solution $c(t)$ to the Becker-Döring equations (1.1), (1.2) for data $c(0)$ such that $\sum_{l=1}^{\infty} l c_l(0) = \rho > \rho_s$, where ρ_s is the maximal density for which an equilibrium solution exists, i.e., $\rho_s = \sum_{l=1}^{\infty} l c_l^s$.

It is easily seen that for coefficients (1.5), (1.6) we have $F(c(0)) < \infty$ if $\sum_{l=1}^{\infty} l c_l(0) < \infty$. Then we have the energy identity

$$F(c(t)) + \int_0^t \sum_{l=1}^{\infty} J_l \ln\left(\frac{a_l c_1 c_l}{b_{l+1} c_{l+1}}\right) ds = F(c(0)), \tag{1.24}$$

and we know that $F(c(t)) \to 0$ as $t \to \infty$.

As in [24] we consider the system for large times when all possible metastable states have broken down. For a small parameter $\varepsilon > 0$ we will in the following use ε^{-1} as a measure for the large clusters. Motivated by the considerations in Section 1.5.1, we will consider times larger than a time t_ε with

$$F(c(t_\varepsilon)) = \varepsilon^{\gamma} \tag{1.25}$$

and introduce a new time scale

$$\tilde{t} = \varepsilon^{1+\gamma-\alpha}(t - t_\varepsilon), \quad \text{for } t \geq t_\varepsilon. \tag{1.26}$$

We define

$$c_l^{\varepsilon}(\tilde{t}) := c_l(t), \quad \text{for } t \geq t_\varepsilon,$$

such that (1.25) reads

$$F(c^{\varepsilon}(0)) = \varepsilon^{\gamma}, \tag{1.27}$$

and (1.1) gives

$$\frac{d}{d\tilde{t}}c_l^\varepsilon = \frac{1}{\varepsilon^{1+\gamma-\alpha}}(J_{l-1}^\varepsilon - J_l^\varepsilon) \tag{1.28}$$

with $J_l^\varepsilon = a_l c_1^\varepsilon c_l^\varepsilon - b_{l+1} c_{l+1}^\varepsilon$ and c_1^ε is such that $\sum_{l=1}^\infty l c_l^\varepsilon(\tilde{t}) = \rho$ for all $\tilde{t}$.

We also need a cut-off $l_0 = l_0(\varepsilon)$ between small and large clusters. For the level of approximation considered in this paper, l_0 only needs to satisfy

$$|c_{l_0}^s|^\eta \;\le\; CF(c^\varepsilon(0)), \quad \text{for any } \eta > 0,$$

$$\lim_{\varepsilon\to 0} l_0^\gamma \sqrt{F(c^\varepsilon(0))} \;=\; 0. \tag{1.29}$$

These requirements are needed to ensure that, on the one hand, l_0 is large enough such that some moment of the rescaled size distribution is bounded (cf. Lemma 2.1), and on the other hand, l_0 is small enough such that the excess density is contained in the clusters larger than l_0 (cf. Lemma 2.2).

For convenience we choose

$$l_0 \approx \frac{1}{\varepsilon^x}, \quad \text{for some } x \in (0, \tfrac{1}{2}). \tag{1.30}$$

Dropping the tilde in the new time scale, we introduce the new variable

$$\lambda = \varepsilon l \tag{1.31}$$

and the rescaled monomer density

$$u^\varepsilon(t) = \varepsilon^{-\gamma}(c_1^\varepsilon(t) - z_s). \tag{1.32}$$

The rescaled densities are defined as measures $\{\nu_t^\varepsilon\}_t \subset C_0^0(\mathbb{R}^+)^*$ via

$$\int_0^\infty \zeta(\lambda)\, d\nu_t^\varepsilon := \frac{1}{\varepsilon}\sum_{l=l_0}^\infty \zeta(\varepsilon l)\, c_l^\varepsilon(t),$$

i.e., on the ε-level ν_t^ε is a sequence of properly rescaled Dirac measures. In the following we will usually omit the integration limits and write $\int \zeta\, d\nu_t^\varepsilon$, $\int \zeta\, d\nu_t$, etc. Later we want to extend the definition to test functions which have not necessarily compact support in $(0,\infty)$. Then, we understand the integral as $\int d\nu_t = \int_{0^+}^\infty d\nu_t$, i.e., the point $\lambda = 0$ is not included.

Our aim is to pass to the limit $\varepsilon \to 0$ and to show that the limits of $\nu^\varepsilon, u^\varepsilon$ satisfy (1.19), (1.20). For that we would like to make as few assumptions as possible on the data, given by $(c_l^\varepsilon(0)) = (c_l(t_\varepsilon))$. It turns out that we only have to make one assumption to ensure that the limit will be nontrivial. We have to assume that at the time when the energy is appropriately small, as in (1.27), not too many very large clusters have formed. More precisely we assume

$$\sum_{l\ge[M/\varepsilon]}^\infty l c_l^\varepsilon(0) = \int_{\lambda\ge M} \lambda\, d\nu_0^\varepsilon \to 0 \quad \text{as } M \to \infty \text{ uniformly in } \varepsilon. \tag{1.33}$$

Theorem 1.1. *Assume that (1.27) and (1.33) hold. Then there exist a subsequence, again denoted by $\varepsilon \to 0$, and a weakly continuous map $[0,\infty) \ni t \mapsto \nu_t \in C_0^0((0,\infty))^*$ such that*

$$\int \zeta \, d\nu_t^\varepsilon \to \int \zeta \, d\nu_t \quad \textit{locally uniform in } t \in \mathbb{R}_+ \quad \textit{for all } \zeta \in C_0^0((0,\infty)).$$

Furthermore there exists $u = u(t) \in L^2_{loc}([0,\infty))$ such that

$$u^\varepsilon(t) \rightharpoonup u \quad \textit{weakly in } L^2_{loc}([0,\infty)).$$

The limit satisfies

$$\partial_t \nu_t + \partial_\lambda \left(\lambda^\alpha \left(u(t) - \frac{q}{\lambda^\gamma} \right) \nu_t \right) = 0 \tag{1.34}$$

in $\mathcal{D}'((0,\infty) \times (0,\infty))$ and

$$\int \lambda \, d\nu_t = \rho - \rho_s, \quad \textit{for all } t \geq 0. \tag{1.35}$$

In addition, if $\alpha \geq 1 - 3\gamma$, we find that (1.35) is equivalent to

$$u(t) = \frac{q \int \lambda^{\alpha-\gamma} \, d\nu_t}{\int \lambda^\alpha \, d\nu_t} \quad \textit{for a.e. } t \geq 0. \tag{1.36}$$

It seems on first glance a bit unfortunate that we cannot conclude (1.36) for all exponents $\alpha, \gamma \in (0,1)$. We are, however, not aware of any example coming from applications where this condition is not satisfied. In particular, it is satisfied for diffusion-controlled as well as interface-reaction-controlled kinetics in two and three dimensions (cf. Section 1.1).

A main ingredient in the proof of Theorem 1.1 is the following estimate, which for the sake of lucidity we state separately.

Proposition 1.2. *The solution ν_t, found in Theorem 1.1, satisfies the following energy estimate: For any $t > 0$, we have*

$$\begin{aligned} \frac{q}{z_s(1-\gamma)} \int \lambda^{1-\gamma} \, d\nu_t + \frac{1}{z_s} \int_0^t \int \lambda^\alpha \left(u(t) - \frac{q}{\lambda^\gamma} \right)^2 d\nu_s \, ds \\ \leq \liminf_{\varepsilon \to 0} \frac{F(c^\varepsilon(0))}{\varepsilon^\gamma} \leq 1. \end{aligned} \tag{1.37}$$

1.5.3. Outline of Proofs and Further Results. The proofs of Theorem 1.1 and Proposition 1.2 are the contents of Section 2.1 and 2.2. Section 2.1 provides several a priori estimates which in principle are all based on the energy estimate given by (1.24). Lemma 2.1 shows how the free energy on the one hand controls the surface energy of the large clusters, i.e., $\int \lambda^{1-\gamma} \, d\nu_t^\varepsilon$, and on the other hand, also gives an estimate of the deviation of the small clusters from their respective equilibrium values c_l^s. Indeed, we will show (cf. (2.2)) that

$$\sum_{l=1}^\infty l^{1-\gamma} |c_l - c_l^s| \leq C\sqrt{F},$$

which replaces the formal argument leading to (1.16) and then to (1.18). This result is used in Lemma 2.2 to conclude that the excess density is contained in the large clusters. In Lemma 2.3 we provide suitable estimates on the fluxes, which can first be used in Lemma 2.4 to show that the tightness property (1.33) is preserved in time.

In Section 2.2 we pass to the limit as $\varepsilon \to 0$. With the estimates from Section 2.1 we easily obtain a weak limit ν_t of ν_t^ε.

The difficulty that remains is to identify the structure of the limit flux, which requires some compactness of the rescaled monomer density u^ε. Unfortunately we cannot use (1.3) to obtain a uniform bound on u^ε. For that it would be necessary to have much better estimates on $\sum_{l=l_0}^{\infty} J_l$ and $\sum_{l=1}^{l_0-1} J_l$ respectively. More precisely, in the limit, the properly rescaled version of the first quantity vanishes, due to the density conservation (1.20). To use (1.3) for a bound on u^ε, we would need a quantitative version of this fact already on the ε-level, which is not available and most likely in general not true. Hence, we will need a different argument in Lemma 2.6 to gain some compactness of u^ε. Finally, Lemma 2.7 provides formula (1.36) for the limit $u(t)$.

In Section 2.3 we investigate further regularity properties of the solution. We will show for a certain range of coefficients that if the data satisfy $\int d\nu_0 < \infty$, then $\int d\nu_t$, i.e., the fraction of clusters remaining at time t is decreasing in time.

An important question is whether the tightness assumption on the data is reasonable. We will discuss this issue in detail in Section 1.5.4 below. In Section 2.4 we will present a sequence of data which is motivated by numerical simulations in [5] and show that this sequence satisfies assumption (1.33) as well as several further regularity properties.

In Section 3 we identify by a formal asymptotic expansion higher-order terms in the evolution of both small and large clusters.

We have seen that to leading order the large clusters satisfy the transport equation (1.19) with characteristics which leave the domain at $\lambda = 0$. That is, there are no boundary conditions which couple the evolution of the large clusters to the evolution of the small clusters, and the large clusters evolve independently of the small ones.

Going one step further in the expansion, the formal asymptotic expansion in Section 3.1 reveals that the small clusters are in quasi-steady equilibrium with the large clusters and uniquely determined by the rescaled monomer density u (cf. equation (3.15)). Then, the density belonging to these small clusters determines a constraint for the next term in an expansion of the large clusters (cf. (3.17)) and so on. Furthermore, we compute in Section 3.2 a detailed asymptotic expansion of the energy estimate through which additional energy-type identities emerge on various levels of approximations (cf. (3.33)–(3.35)).

Let us mention here another rigorous study of the transition from the Becker-Döring equations and (1.19), which is complementary to ours. In [15], the case of homogeneous coefficients is studied, i.e., (1.5), (1.6) with the assumption $z_s = 0$ and $\alpha \geq \gamma$, or in other words, the case where coagulation dominates fragmentation. In this case there exists no nontrivial equilibrium and no useful Lyapunov functional; in [6] weak convergence of the solution to zero is shown. Thus, in this case the total density ρ is contained in the large clusters, or in other words, all clusters are large clusters, and one can in this special case use (1.3) to derive a uniform bound on u^ε.

In principle, the method we developed here can be directly applied to generalized Becker-Döring models for which a bounded Lyapunov functional exists—for example, Becker-Döring models which allow for multiple components (see e.g. [11] and [9]) or which take into account the autocatalytic production of monomers [8]. For a different scaling limit of the Becker-Döring equations for uniformly bounded coefficients we also refer to [7].

1.5.4. Remarks on the Data for the Large-Time Regime and a Review of Coarsening Dynamics. We obtain under the single assumption (1.33) on the data that the large clusters are in the long-time limit described by (1.19). One might wonder whether assumption (1.33) is a natural one. Of course, one could easily construct a sequence of data with small energy but violating (1.33). The question of interest is, however, whether the Becker-Döring equations can create such data for the time regime of interest here, starting from generic data, e.g. only monomers.

The problem in analyzing the long-time behavior of the Becker-Döring system is the appearance of metastable states which have been described in Section 1.3. Since we expect that any solution typically goes through a metastable state, the task would be to analyze how the solution leaves this metastable state. It is hard to imagine a mechanism, which, at least for data which are for example only monomers, creates very large clusters in the sense that (1.33) would be violated; numerical simulations in [5] confirm this, but a proof is presently not available.

However, we construct in Section 2.4 a sequence of data which is motivated by the simulations in [5]. A striking similarity is observed between a solution of the Becker-Döring equations going through a metastable state and a sequence of equilibrium solutions for a finite system of size n, letting $n \to \infty$. We use the latter to construct a sequence of data for the regime under consideration. Indeed, we can show that these data behave nicely in an appropriate sense and, in particular, satisfy assumption (1.33).

To understand what type of data can be created by the Becker-Döring equations is also of interest for another reason, which concerns the large-time behavior of solutions to (1.19), (1.21) and is related to the classical LSW theory of coarsening. Let us explain this issue briefly, since it is a major motivation for studying the connection of the Becker-Döring equations and their scaling limit.

For this discussion let us concentrate on the specific case of diffusion-controlled cluster growth in three dimensions, i.e., coefficients with $\alpha = \gamma = 1/3$, which leads to (1.17), (1.18), the classical LSW model. The qualitative features we describe now for this specific case are the same for solutions of equation (1.19), (1.21) with general coefficients. The LSW model describes the large-time behavior of the stable nuclei in a phase transition in the regime of small volume fraction of the new phase. The driving force in this last stage of the phase transition is the surface energy, and to reduce this surface energy, clusters interact by diffusional mass exchange, such that atoms diffuse from small to large clusters. Thus, large clusters grow, and smaller ones shrink and disappear, a process which leads to an increase in the typical length scales in the system, i.e., to coarsening of the microstructure. This form of competitive growth is also known as Ostwald Ripening and is a fundamental process in the aging of materials (see [32] for a review on the physical background). The LSW model can also be derived as a homogenization limit of the Mullins-Sekerka free boundary problem [17], [18].

As part of their classical theory, LSW predicted that the large-time behavior of solutions to (1.17), (1.18) is universal and characterized by a smooth self-similar solution with compact support. As a consequence, one obtains universal power laws for averaged quantities, characterizing the dynamics of a system undergoing Ostwald Ripening. However, the LSW theory has a major drawback. For equation (1.17), (1.18) there exists a one-parameter family of self-similar solutions, and a rigorous analysis in [19], [21] for data with compact support shows that the long-time behavior is not at all universal but depends, on the contrary, very sensitively on the data. More precisely, it depends on the detailed behavior of the initial distribution of large clusters, i.e., on the details at the end of the support. Indeed, the size distribution approaches the particular self-similar solution, which displays the same behavior at the end of its support (for details see [19], [21]).

The question is now: What are natural modifications to overcome this nonphysical weak selection criterion of asymptotic states?

In [31], the formal relation between the Becker-Döring and the LSW model is used, but the second-order term represented by $b_{l+1}c_{l+1} - b_l c_l$, (cf. (1.12)) is kept, and it is argued for a corresponding continuous partial differential equation that the solution predicted by LSW is the only possible limit in self-similar variables. The main effect of this additional term is to create a fast-decaying infinite tail for compactly supported data.

Another argument in favor of universal self-similar asymptotics would be that the data for the coarsening regime exhibit a certain universal behavior. Even though the Becker-Döring equations can only give a simplified picture of the subtle phenomenon of nucleation, having established the connection with the LSW model and the sensitivity of the LSW model with respect to the data, it would be very interesting to understand which data are typically created by the Becker-Döring dynamics. As we have explained before, the existence of metastable states indicates that a rigorous answer to this question will be difficult to obtain.

2. Rigorous Derivation of Leading Order Dynamics

2.1. A Priori Estimates

In this section and in Section 2.2 we provide the proofs of Theorem 1.1 and Proposition 1.2 respectively.

In the following we will for convenience drop the superscript ε in c_l^ε and J_l^ε. Recall that due to (1.27),

$$F(c(t)) + \frac{1}{\varepsilon^{1+\gamma-\alpha}} \int_0^t \sum_{l=1}^{\infty} J_l \ln\left(\frac{a_l c_1 c_l}{b_{l+1} c_{l+1}}\right) ds = F(c(0)) = \varepsilon^\gamma, \tag{2.1}$$

for all $t \in [0, \infty)$.

We start with some a priori bounds which follow from the convexity properties of f as in (1.23), the first being a simple version of the so-called Csiszar-Kullback inequality. In the following C will denote a generic constant which in general will depend on the parameters α, γ, q, z_s, and ρ.

Lemma 2.1. *If (1.27) holds, then we have for all $t \geq 0$,*

$$\sum_{l=1}^{\infty} l^{1-\gamma} |c_l(t) - c_l^s| \;\leq\; C\sqrt{F(c(t))} \leq C\varepsilon^{\gamma/2}, \tag{2.2}$$

$$\frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} c_l(t) \;\leq\; \tfrac{1}{1-\eta} F(c(t)) + C_{\eta,p}\varepsilon^p, \tag{2.3}$$

for any small $\eta > 0$ and any $p < \infty$.

Proof. We use the duality relation

$$yz \leq f(z) + f^*(y), \tag{2.4}$$

where f^* is the dual of f and is given by

$$f^*(y) = e^y - y - 1. \tag{2.5}$$

Notice that f and f^* satisfy

$$f(|z|) \leq f(z) \qquad \text{and} \qquad f^*(ry) \leq r^2 f^*(y), \quad \text{for } r \in [0,1]. \tag{2.6}$$

With $y = \delta(1-\eta)\frac{q}{(1-\gamma)z_s} l^{1-\gamma}$ for some $\delta, \eta \in (0,1]$ and $z = \frac{c_l - c_l^s}{c_l^s}$, we find with (2.4), (2.5), and (2.6) that

$$\delta \frac{(1-\eta)q}{(1-\gamma)z_s} l^{1-\gamma} \frac{|c_l - c_l^s|}{c_l^s} \leq \delta^2 \exp\left\{\frac{(1-\eta)q}{(1-\gamma)z_s} l^{1-\gamma}\right\} \,+\, f\left(\frac{c_l - c_l^s}{c_l^s}\right).$$

If we multiply with c_l^s, sum over $l \geq L$, and use the fact that due to (1.8) it follows that $c_l^s \leq C\exp\{-\frac{q}{(1-\gamma)z_s} l^{1-\gamma}\}$, we find

$$\begin{aligned}\frac{q}{(1-\gamma)z_s} \sum_{l=L}^{\infty} l^{1-\gamma} |c_l - c_l^s| \;\leq\; & \frac{C\delta}{1-\eta} \sum_{l=L}^{\infty} \exp\left\{-\frac{\eta q}{(1-\gamma)z_s} l^{1-\gamma}\right\} \\ & + \frac{1}{\delta(1-\eta)} F(c).\end{aligned}$$

Now we observe that

$$\begin{aligned}\sum_{l=L}^{\infty} \exp\left\{-\frac{\eta q}{(1-\gamma)z_s} l^{1-\gamma}\right\} \;\leq\; & C \int_L^{\infty} \exp\left\{-\frac{\eta q}{(1-\gamma)z_s} l^{1-\gamma}\right\} dl \\ \leq\; & C_\eta \exp\left\{-\frac{\eta q}{(1-\gamma)z_s} L^{1-\gamma}\right\}.\end{aligned}$$

Hence, if we choose $L = 1$, $\delta = \sqrt{F}$, and $\eta = \frac{1}{2}$, for example, we find (2.2). On the other hand, if we choose $\delta = 1$ and $L = l_0$ and recall (1.30), we obtain

$$\frac{q}{z_s(1-\gamma)} \sum_{l=l_0}^{\infty} l^{1-\gamma} |c_l - c_l^s| \leq \frac{1}{1-\eta} F(c(t)) + C_{\eta,p} O(\varepsilon^p), \tag{2.7}$$

for any $p < \infty$. Since

$$\int_L^\infty l^x e^{-c_0 l^{1-\gamma}}\, dl \sim L^{x+\gamma} e^{-c_0 L^{1-\gamma}}, \tag{2.8}$$

it follows

$$\sum_{l=l_0}^\infty l^{1-\gamma} c_l^s \le C_p \varepsilon^p$$

for any $p < \infty$, which together with (2.7) proves (2.3). □

Now we are in the position to prove that the excess density is almost completely contained in the clusters which are larger than l_0.

Lemma 2.2. *Under the assumption (1.27) we find for all $t \ge 0$,*

$$\left| \sum_{l=l_0}^\infty l c_l(t) - (\rho - \rho_s) \right| \le C\, l_0^\gamma \sqrt{F(c(t))} + C_p \varepsilon^p \le C \varepsilon^{\gamma(1/2 - x)},$$

with $p < \infty$ and x as in (1.30).

Proof. We conclude with (1.2), (1.9), (1.30), (2.2), and (2.8) that

$$\begin{aligned} \left| \sum_{l=l_0}^\infty l c_l(t) - (\rho - \rho_s) \right| &= \sum_{l=1}^{l_0 - 1} l (c_l - c_l^s) + \sum_{l=l_0}^\infty l c_l^s \\ &\le C l_0^\gamma \sqrt{F(c(t))} + C_p \varepsilon^p. \end{aligned}$$
□

In the following lemma we use the bound on the energy dissipation rate to derive a bound on the flux J_l.

Lemma 2.3. *Under the assumption (1.27) the following inequalities hold:*

$$\int_0^\infty \sum_{l=1}^\infty \frac{|J_l|^2}{\max(a_l c_1 c_l, b_{l+1} c_{l+1})}\, dt \le \varepsilon^{1+\gamma-\alpha} F(c(0)) = \varepsilon^{1+2\gamma-\alpha}, \tag{2.9}$$

$$\int_0^\infty \eta(t) \sum_{l=l_0}^\infty l^\kappa |J_l|\, dt \le C\, \|\eta\|_{L^2((0,\infty))} \varepsilon^{1-\alpha+\gamma-\kappa}, \tag{2.10}$$

for $\eta \in L^2((0,\infty))$ and $\kappa \in \left[\frac{1-\alpha-\gamma}{2}, \frac{1-\alpha}{2}\right]$.

Proof. Inequality (2.9) is a direct consequence of (1.27), (2.1) and the fact that $(x-y)\ln\left(\frac{x}{y}\right) \ge \frac{(x-y)^2}{\max(x,y)}$.

To see (2.10) we first use Cauchy-Schwarz's inequality and recall (1.5), (1.6) to get

$$\sum_{l=l_0}^{\infty} l^{\kappa}|J_l| \leq C\left(\sum_{l=1}^{\infty}\frac{|J_l|^2}{\max(a_l c_1 c_l, b_{l+1}c_{l+1})}\right)^{1/2}\left(\sum_{l=l_0}^{\infty} l^{2\kappa+\alpha}c_l\right)^{1/2}.$$

Now we use Hölder's inequality to find

$$\sum_{l=l_0}^{\infty} l^{2\kappa+\alpha}c_l \leq \left(\sum_{l=l_0}^{\infty} l c_l\right)^{x_1}\left(\sum_{l=l_0}^{\infty} l^{1-\gamma}c_l\right)^{x_2},$$

with

$$x_1 = \frac{2\kappa+\alpha+\gamma-1}{\gamma} \qquad \text{and} \qquad x_2 = \frac{1-2\kappa-\alpha}{\gamma},$$

with the requirement that $1-\alpha \geq 2\kappa \geq 1-\alpha-\gamma$. Summarizing these inequalities and using (1.2), (2.3), and (2.9) we find

$$\int_0^{\infty}\eta(t)\sum_{l=l_0}^{\infty} l^{\kappa}|J_l|\,ds \leq C\|\eta\|_{L^2((0,\infty))}\,\varepsilon^{(1-\alpha)/2+\gamma}\,\varepsilon^{(1-2\kappa-\alpha)/2},$$

which finishes the proof of the Lemma. □

The bound on the flux now enables us to show that the tightness property (1.33) is preserved in time.

Lemma 2.4. *If (1.27) and (1.33) hold, then for all $t \geq 0$,*

$$\sum_{l=[M/\varepsilon]}^{\infty} l c_l(t) \to 0, \quad \text{as } M \to \infty \quad \text{uniformly in } \varepsilon > 0.$$

Proof. Let $N \gg M \gg 1$ and let $\phi \in C^1(\mathbb{R}^+)$ be a cut-off function such that $\phi(l) = 0$ for $l \leq \frac{M}{2\varepsilon}$ and $l \geq \frac{2N}{\varepsilon}$, $\phi(l) = 1$ for $\frac{M}{\varepsilon} \leq l \leq \frac{N}{\varepsilon}$ and such that $\phi' \leq C\frac{\varepsilon}{M}$ for $l \in (\frac{M}{2\varepsilon}, \frac{M}{\varepsilon})$ and $|\phi'| \leq \frac{\varepsilon}{N}$ for $l \in (\frac{N}{\varepsilon}, \frac{2N}{\varepsilon})$. Then we find using (1.2),

$$\begin{aligned}
\frac{d}{dt}\sum_{l=1}^{\infty}\phi(l) l c_l(t) &= -\frac{1}{\varepsilon^{1-\alpha+\gamma}}\sum_{l=1}^{\infty}\phi(l) l (J_l - J_{l-1}) \\
&= \frac{1}{\varepsilon^{1-\alpha+\gamma}}\sum_{l=1}^{\infty}(\phi(l+1)(l+1) - \phi(l) l) J_l \\
&= \frac{1}{\varepsilon^{1-\alpha+\gamma}}\left(\sum_{l=1}^{\infty}\phi(l+1)J_l + \sum_{l=1}^{\infty}(\phi(l+1)-\phi(l)) l J_l\right) \\
&\leq \frac{C}{\varepsilon^{1-\alpha+\gamma}}\left(\sum_{l\geq[M/2\varepsilon]}|J_l| + \frac{\varepsilon}{M}\sum_{l=[M/2\varepsilon]}^{[M/\varepsilon]} l|J_l| + \frac{\varepsilon}{N}\sum_{l=[N/\varepsilon]}^{[2N/\varepsilon]} l^{1+\alpha}c_l\right)
\end{aligned}$$

$$\le \frac{C}{\varepsilon^{1-\alpha+\gamma}} \left(\left(\frac{\varepsilon}{M}\right)^{(1-\alpha)/2} \sum_{l=l_0}^{\infty} l^{(1-\alpha)/2} |J_l| \right.$$
$$\left. + \frac{\varepsilon}{M} \left(\frac{M}{\varepsilon}\right)^{1-(1-\alpha)/2} \sum_{l=l_0}^{\infty} l^{(1-\alpha)/2} |J_l| + \left(\frac{\varepsilon}{N}\right)^{1-\alpha} \right)$$
$$\le \frac{C}{M^{(1-\alpha)/2}} \varepsilon^{-(1-\alpha)/2-\gamma} \sum_{l=l_0}^{\infty} l^{(1-\alpha)/2} |J_l| + C \frac{\varepsilon^{-\gamma}}{N^{1-\alpha}}.$$

Integrating over time and using (2.10) for $\kappa = (1-\alpha)/2$, we find

$$\sum_{l \ge [M/\varepsilon]}^{[N/\varepsilon]} l c_l(t) \le \sum_{l \ge [M/2\varepsilon]}^{[N/\varepsilon]} l c_l(0) + C \frac{t^{1/2}}{M^{(1-\alpha)/2}} + C \frac{t \varepsilon^{-\gamma}}{N^{1-\alpha}}.$$

If we let N tend to ∞, the assertion of the Lemma follows by (1.33). □

2.2. Passage to the Limit

For the following we define the rescaled fluxes as signed measures $\{\mu_t^\varepsilon\}_t \subset C_0^0(\mathbb{R}^+)^*$ by

$$\int \zeta(\lambda)\, d\mu_t^\varepsilon := \frac{1}{\varepsilon^{1+\gamma-\alpha}} \sum_{l=l_0}^{\infty} \zeta(\varepsilon l)\, J_l(t). \tag{2.11}$$

For later purposes we also define

$$\mathcal{D}^\varepsilon := \frac{1}{\varepsilon^{1-\alpha+2\gamma}} \sum_{l=l_0}^{\infty} J_l \ln\left(\frac{a_l c_1 c_l}{b_{l+1} c_{l+1}}\right), \tag{2.12}$$

and note that due to (2.1) the inequality

$$\int_0^\infty \mathcal{D}^\varepsilon(t)\, dt \le 1 \tag{2.13}$$

holds.

Then the a priori estimates in Lemmas 2.1–2.4 translate into the following bounds for ν_t^ε and μ_t^ε respectively:

$$\sup_{t\in(0,\infty)} \int \lambda^{1-\gamma}\, d\nu_t^\varepsilon \le C, \tag{2.14}$$

$$\sup_{t\in(0,\infty)} \left| \int \lambda\, d\nu_t^\varepsilon - (\rho - \rho_s) \right| \le C \varepsilon^{\gamma(1/2-x)}, \tag{2.15}$$

$$\sup_{\varepsilon\to 0} \int_M^\infty \lambda\, d\nu_t^\varepsilon \to 0, \quad \text{as } M \to \infty \text{ for all } t \ge 0, \tag{2.16}$$

$$\int_0^\infty \eta(t) \int \lambda^\kappa |d\mu_t^\varepsilon|\, dt \le C \|\eta\|_{L^2((0,\infty))} \tag{2.17}$$
$$\text{for } \kappa \in \left[\frac{1-\alpha-\gamma}{2}, \frac{1-\alpha}{2}\right].$$

Furthermore we immediately conclude with (2.10) the following weak Hölder regularity of ν_t^ε. For all $t_1, t_2 \in [0,\infty)$ and all $\zeta \in C_0^1((0,\infty))$, we find for $\tilde{l} \in [l, l+1]$,

$$\begin{aligned}
\left| \int \zeta \, d\nu_{t_1}^\varepsilon - \int \zeta \, d\nu_{t_2}^\varepsilon \right| &= \frac{1}{\varepsilon^{1-\alpha+\gamma}} \int_{t_1}^{t_2} \frac{d}{dt} \sum_{l=l_0}^{\infty} \zeta(\varepsilon l)\, c_l(t)\, dt \\
&\le \frac{1}{\varepsilon^{1-\alpha+\gamma}} \int_{t_1}^{t_2} \sum_{l=l_0}^{\infty} \zeta'(\varepsilon \tilde{l})\, |J_l(t)|\, dt \\
&\le \frac{1}{\varepsilon^{1-\alpha+\gamma}} \sup_\lambda \frac{|\zeta'(\lambda)|}{\lambda^{(1-\alpha)/2}} \varepsilon^{(1-\alpha)/2} \int_{t_1}^{t_2} \sum_{l=l_0}^{\infty} l^{(1-\alpha)/2} |J_l(t)|\, dt \\
&\le C \sup_\lambda \frac{|\zeta'(\lambda)|}{\lambda^{(1-\alpha)/2}} |t_1 - t_2|^{1/2}. \qquad (2.18)
\end{aligned}$$

The uniform bounds (2.14) and (2.18) ensure with Arzela-Ascoli that there exists a weakly continuous family $\{\nu_t\}_t$ of nonnegative Borel measures on $(0,\infty)$ such that for a subsequence

$$\int \zeta \, d\nu_t^\varepsilon \to \int \zeta \, d\nu_t, \quad \text{locally uniform in } t \in [0,\infty), \qquad (2.19)$$

for ζ in a countable subset of $C_0^1((0,\infty))$. Again by (2.14), (2.15), and (2.16) we find that we can extend the convergence in (2.19) to all $\zeta \in C^0((0,\infty))$ which satisfy

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

and this implies

$$\int \lambda \, d\nu_t = \rho - \rho_s, \quad \text{for all } t \in [0,\infty). \qquad (2.20)$$

In addition, the bounds (2.14) and (2.15) give by weak lower semicontinuity that

$$\sup_{t\in(0,\infty)} \frac{q}{z_s(1-\gamma)} \int \lambda^{1-\gamma} \, d\nu_t \le 1. \qquad (2.21)$$

For the fluxes (2.17) ensures that there exists a signed measure $\mu \in C_0^0((0,\infty)\times(0,\infty))^*$ such that for a further subsequence

$$\int\int \xi(\lambda,t)\, d\mu_t^\varepsilon \, dt \to \int\int \xi \, d\mu, \qquad (2.22)$$

for all $\xi \in C_0^0((0,\infty)\times(0,\infty))$. The following lemma shows that μ is absolutely continuous with respect to the limit density.

Lemma 2.5. *There exists a function $v \in L^2(\lambda^{-\alpha}\, d\nu_t\, dt)$ such that*

$$\int\int \xi(\lambda,t)\, d\mu = \int\int \xi(\lambda,t)\, v(\lambda,t)\, d\nu_t\, dt,$$

and for all $t > 0$ we find

$$\frac{1}{z_s}\int_0^t \int \frac{|v|^2}{\lambda^\alpha}\, d\nu_s\, ds \le \liminf_{\varepsilon\to 0}\int_0^t \mathcal{D}^\varepsilon(s)\, ds. \tag{2.23}$$

Proof. We introduce $d\tilde{\mu}_t^\varepsilon\, dt = \lambda^{-(1+\alpha)/2} d\mu_t^\varepsilon\, dt$. Due to (2.22) there exists for a subsequence a weak$*$ limit $d\tilde{\mu} = \lambda^{-(1+\alpha)/2} d\mu$. With (2.9) we obtain for any $T < \infty$ and $\xi \in C_0^0((0,T)\times(0,\infty))$,

$$\begin{aligned}
\int\int \xi\lambda\, d\tilde{\mu}_t^\varepsilon\, dt &= \int\int \xi\lambda^{(1-\alpha)/2}\, d\mu_t^\varepsilon\, dt\\
&= \varepsilon^{(\alpha-1)/2-\gamma}\int\sum_{l=l_0}^\infty \xi l^{(1-\alpha)/2} J_l(t)\, dt\\
&\le \left(\int_0^T \mathcal{D}^\varepsilon(t)\, dt\right)^{1/2}\\
&\quad\cdot\left(\int\sum_{l=l_0}^\infty \xi^2 l^{1-\alpha}\max(a_l c_1 c_l, b_{l+1}c_{l+1})\, dt\right)^{1/2}\\
&= \left(\int_0^T \mathcal{D}^\varepsilon(t)\, dt\right)^{1/2}\\
&\quad\cdot\left(\max\left(c_1\int\sum_{l=l_0}^\infty \xi^2 l c_l, \int\sum_{l=l_0}^\infty \xi^2 l\left(z_s+\frac{q}{l^\gamma}\right)c_l\right)\right)^{1/2}.
\end{aligned}$$

The right-hand side is uniformly bounded due to (1.2). We take a subsequence $\varepsilon \to 0$ such that

$$\int_0^T \mathcal{D}^\varepsilon(t)\, dt \to \liminf_{\varepsilon\to 0}\int_0^T \mathcal{D}^\varepsilon(t)\, dt =: K,$$

and we find for fixed ξ that

$$\int\int \xi\lambda d\tilde{\mu}_t\, dt \le \left(K z_s\int\int \xi^2\lambda\, d\nu_t\, dt\right)^{1/2}.$$

Since continuous functions with compact support are dense in $L^2(\lambda d\nu_t\, dt)$, there exists by Riesz a function $\tilde{v} \in L^2(\lambda d\nu_t\, dt)$ such that

$$\int\int \xi\lambda\, d\tilde{\mu}\, dt = \int\int \xi\tilde{v}\lambda\, d\nu_t\, dt,$$

and we have

$$\int\int \xi\lambda\tilde{v}\, d\nu_t\, dt \le \left(K z_s\int\int \xi^2\lambda\, d\nu_t\, dt\right)^{1/2}. \tag{2.24}$$

With $v = \lambda^{(1+\alpha)/2}\tilde{v}$ the first part of the lemma is proved. By approximation we can also take $\xi = \frac{v}{z_s\lambda^{(1+\alpha)/2}}$ in (2.24), and the second part follows. □

From now on we fix a subsequence $\varepsilon \to 0$ such that the convergence in (2.19) and (2.22) holds. Notice that

$$\partial_t \nu_t + \partial_\lambda (v\, \nu_t) = 0 \tag{2.25}$$

holds in the sense of distributions.

The next lemma provides weak convergence in L^2, and consequently a bound, for $u^\varepsilon(t)$ and it identifies the structure of v.

Lemma 2.6. *There exists $u \in L^2_{loc}([0,\infty))$ such that*

$$u^\varepsilon \rightharpoonup u, \quad \text{in } L^2_{loc}([0,\infty)), \tag{2.26}$$

and

$$v(\lambda,t) = \lambda^\alpha \left(u(t) - \frac{q}{\lambda^\gamma} \right), \quad \text{for a.a. } t \text{ and } \nu_t - \text{a.e. in } \mathbb{R}^+. \tag{2.27}$$

Proof. We choose for any given $T < \infty$,

$$\eta = \eta(t) \in L^2((0,T)) \qquad \text{and} \qquad \zeta = \zeta(\lambda) \in C^1_0((0,\infty)).$$

We will show

$$\int_0^T \eta \int \zeta \, d\mu_t^\varepsilon \, dt = \int_0^T \eta \left(u^\varepsilon \int \zeta \lambda^\alpha \, d\nu_t^\varepsilon - q \int \zeta \lambda^{\alpha-\gamma} \, d\nu_t^\varepsilon \right) dt + \int_0^T \eta \omega(\zeta, t, \varepsilon) \, dt, \tag{2.28}$$

with

$$\sup_{t \in (0,\infty)} |\omega| \le C \sup_{\lambda \in (0,\infty)} |\zeta'(\lambda)| \, \varepsilon^{1-\gamma} \left(\int_{\mathrm{supp}(\zeta)} \lambda^\alpha \, d\nu_t^\varepsilon + \varepsilon^\gamma \int_{\mathrm{supp}(\zeta)} \lambda^{\alpha-\gamma} \, d\nu_t^\varepsilon \right).$$

Notice that, once we have proved (2.28) as well as (2.26), the formula (2.27) follows directly. To prove (2.28) we recall with (1.5), (1.6), and (2.11) that

$$\begin{aligned}
\int \zeta \, d\mu_t^\varepsilon &= \frac{1}{\varepsilon^{1-\alpha+\gamma}} \sum_{l=l_0}^\infty \zeta J_l \\
&= \frac{1}{\varepsilon^{1-\alpha+\gamma}} \sum_{l=l_0}^\infty \zeta a_l \left((c_1 - z_s) - \frac{q}{l^\gamma} \right) c_l(t) \\
&\quad - \frac{1}{\varepsilon^{1-\alpha+\gamma}} \sum_{l=l_0}^\infty \zeta (b_{l+1} c_{l+1} - b_l c_l).
\end{aligned}$$

Now, since ζ has compact support,

$$\sum_{l=l_0}^\infty \zeta(\varepsilon l)(b_{l+1}c_{l+1} - b_l c_l) = \sum_{l=l_0}^\infty (\zeta(\varepsilon(l-1)) - \zeta(\varepsilon l)) b_l c_l,$$

and hence taking into account (1.6) and (1.31),

$$\int \zeta \, d\mu_t^\varepsilon = \int \zeta \lambda^\alpha \Big(u^\varepsilon(t) - \frac{q}{\lambda^\gamma} \Big) \, d\nu_t^\varepsilon - \frac{1}{\varepsilon^\gamma} \int (\zeta(\lambda - \varepsilon) - \zeta(\lambda)) \lambda^\alpha \Big(z_s + \frac{q \varepsilon^\gamma}{\lambda^\gamma} \Big) \, d\nu_t^\varepsilon.$$

Then (2.28) follows, since $|\zeta(\lambda - \varepsilon) - \zeta(\lambda)| \leq \varepsilon \sup_\lambda |\zeta'|$.

We know by (2.19) that

$$z^\varepsilon(t) := \int \zeta \lambda^\alpha \, d\nu_t^\varepsilon \to \int \zeta \lambda^\alpha \, d\nu_t =: z(t),$$

and

$$\int \zeta \lambda^{\alpha - \gamma} \, d\nu_t^\varepsilon \to \int \zeta \lambda^{\alpha - \gamma} \, d\nu_t$$

locally uniform in $t \in [0, \infty)$. Furthermore, due to (2.20) we can find for all $T < \infty$ a ζ and a constant $c_T > 0$ such that

$$z(t) \geq c_T \qquad \text{and consequently} \qquad z^\varepsilon(t) \geq \frac{c_T}{2}, \tag{2.29}$$

for $t \leq T$ and for sufficiently small ε. For $\eta \in C^0([0, T])$ we find with (2.28) that

$$\int \eta u^\varepsilon z^\varepsilon \, dt = \int \int \eta \zeta \, d\mu_t^\varepsilon \, dt + q \int \int \eta \zeta \lambda^{\alpha - \gamma} \, d\nu_t^\varepsilon \, dt + \int \int \eta \omega \to \int \int \eta \zeta v \, d\nu_t \, dt + q \int \int \eta \zeta \lambda^{\alpha - \gamma} \, d\nu_t \, dt.$$

Since $C_0^0([0, T))$ is dense in $L^2((0, T))$ and $v \in L^2(\lambda^{-\alpha} \, d\nu_t \, dt)$, the bound (2.10) also ensures that the above convergence holds for $\eta \in L^2((0, T))$.

Hence, $u^\varepsilon z^\varepsilon \to \tilde{z}$ in $L^2_{loc}([0, \infty))$. Since $z^\varepsilon \to z$ uniformly and due to (2.29), the assertion of the lemma follows. □

The energy estimate (1.37) is now an immediate consequence of (2.1), (2.3), (2.23), and (2.27).

We still do not have an explicit expression for $u(t)$. Formally, we conclude from the equation for ν_t and density conservation (2.15) that (1.36) holds. However, we do not a priori know whether the lower moments are finite. One can show that the moments are finite for almost all times, but we omit the proof here since it is not very useful to proceed further.

With some additional assumptions on α and γ (which include all cases interesting for applications), we can conclude that these moments are integrable in time and (1.36) holds for almost all times.

Lemma 2.7. *If $\alpha \geq 1 - 3\gamma$, the formula*

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

holds for almost all $t > 0$.

Proof. We first note

$$\int\int |v|\,d\nu_t\,dt \le \left(\int\int \frac{|v|^2}{\lambda^\alpha}\,d\nu_t\,dt\right)^{1/2}\left(\int\int \lambda^\alpha\,d\nu_t\,dt\right)^{1/2}. \tag{2.30}$$

Let us for the moment assume that

$$\int \lambda^\alpha\,d\nu_t \;\in L^1((0,T)), \tag{2.31}$$

for any $T < \infty$. Then, due to (2.30) and density conservation (2.20), we can conclude that $\int v\,d\nu_t = 0$ for almost all t. This follows by an approximation argument: For any $0 \le t_1 < t_2 < \infty$ and a suitable cut-off function $\zeta_\delta(\lambda)$, we find with (2.25),

$$\int_{t_1}^{t_2} \frac{d}{dt}\int \zeta_\delta \lambda\,d\nu_t\,dt = \int_{t_1}^{t_2}\int \zeta_\delta v\,d\nu_t\,dt + \int_{t_1}^{t_2}\int \zeta_\delta' \lambda v\,d\nu_t\,dt. \tag{2.32}$$

Now the last term on the right-hand side can be estimated as

$$\begin{aligned}\int_{t_1}^{t_2}\int \zeta_\delta'\lambda v\,d\nu_t\,dt \;&\le\; \int_{t_1}^{t_2}\left[\frac{1}{\delta}\int_\delta^{2\delta}\lambda|v|\,d\nu_t + \delta\int_{1/\delta}^{2/\delta}\lambda|v|\,d\nu_t\right]dt\\ &\le\; C\int_{t_1}^{t_2}\left[\int_\delta^{2\delta}|v|\,d\nu_t + \int_{1/\delta}^{2/\delta}|v|\,d\nu_t\right]dt\\ &\to\; 0, \quad \text{as } \delta \to 0.\end{aligned}$$

Since the term on the left-hand side of (2.32) converges to zero as $\delta \to 0$ due to (2.20) and the first term on the right-hand side converges to $\int\int v\,d\nu_t\,dt$ due to the integrability of v, we find $\int v\,d\nu_t = 0$ for almost all t. The formula for u follows then from (2.27).

Let us now investigate when (2.31) holds. We first note that if $\alpha \ge 1-\gamma$, it follows from (2.20) and (2.21) that $\int \lambda^\alpha\,d\nu_t \in L^\infty((0,\infty))$. Otherwise we can conclude that $\int \lambda^{1-\gamma-\alpha} v\,d\nu_t \in L^2((0,T))$. This follows from

$$\begin{aligned}\int_0^T\left(\int_{(0,1)}\lambda^{1-\gamma-\alpha}v\,d\nu_t\right)^2 dt \;&\le\; \int\left(\int\frac{|v|^2}{\lambda^\alpha}\,d\nu_t\;\int_{(0,1)}\lambda^{2-2\gamma-\alpha}\,d\nu_t\right)dt\\ &\le\; C\sup_{t\in(0,T)}\int_{(0,1)}\lambda^{2-2\gamma-\alpha}\,d\nu_t,\end{aligned}$$

which is bounded if $1-\gamma \ge \alpha$. Hence, due to $v = \lambda^\alpha(u(t) - \frac{q}{\lambda^\gamma})$, we find $\int \lambda^{1-2\gamma}\,d\nu_t \in L^2((0,T))$, and consequently $\int \lambda^\alpha\,d\nu_t \in L^2((0,T))$ if $\alpha \ge 1-2\gamma$.

Furthermore,

$$\int_0^T\left|\int_{(0,1)}\lambda^{1-2\gamma-\alpha}v\,d\nu_t\right| dt \le \left(\int\int\frac{|v|^2}{\lambda^\alpha}\,d\nu_t\,dt\right)^{1/2}\left(\int\int_{(0,1)}\lambda^{2-4\gamma-\alpha}\,d\nu_t\,dt\right)^{1/2}.$$

The second factor is bounded for $\alpha \le 1-2\gamma$, and we find in this case $\int \lambda^{1-3\gamma}\,d\nu_t \in L^1((0,T))$, which finishes the proof of the lemma. □

2.3. Further Regularity

In this section we show that for a certain range of coefficients the total number density is indeed decreasing for all times.

Let us explain the general difficulties we face if we want to prove such a statement. Naively, we would expect in the case $\alpha \geq \gamma$ that, even if $\int d\nu_0$ is not finite, after an initial time layer, the mass sitting at zero would be transported to the left and afterwards $\int d\nu_t$ would decrease. This in particular would imply that $u(t)$ is bounded for all $t > 0$. Slightly less ambitious, one would like to show that this scenario is true, if $\int d\nu_0 < \infty$. For this setting, uniqueness of solutions has been shown in [22] (see also [14] for a study of L^1-solutions). As a consequence one obtains the following: If for a sequence $\varepsilon \to 0$, it is satisfied that $\nu_0^\varepsilon \rightharpoonup \nu_0$, we have $\nu_t^\varepsilon \rightharpoonup \nu_t$ for all t, and ν_t is the unique solution of (1.34), (1.35).

However, the proof that $\int d\nu_t$ is indeed decreasing turns out to be difficult. The main reason is that $\int d\nu_t$ is not necessarily continuous in time, so we cannot use a bootstrap-type argument. Another strategy is to regularize either ν_t or $u(t)$, but then we have to prove that the regularized solution converges to the original one, which either requires a uniqueness result for the limit equation, which is not available for $u \in L^2((0, T))$, or a strong type of convergence. The following Lemma 2.8 shows that we can prove the latter for a certain range of coefficients.

Before we proceed, we remark that another approach to improve the regularity of the solution would be to rigorously justify an additional energy-type estimate. Indeed, one easily checks that formally the following formula is satisfied for $\gamma > 1/2$:

$$\frac{d}{dt}\left(\frac{q^2}{z_s^2(1-2\gamma)}\int \lambda^{1-2\gamma}\, d\nu_t\right) = -\frac{u}{z_s^2}\int \frac{(\lambda^\alpha u - q\lambda^{\alpha-\gamma})^2}{\lambda^\alpha}\, d\nu_t - \frac{q}{z_s^2}\int \frac{(\lambda^\alpha u - q\lambda^{\alpha-\gamma})^2}{\lambda^{\alpha+\gamma}}\, d\nu_t.$$

A rigorous justification would give additional bounds on lower moments by proceeding as in the proof of Lemma 2.7.

Lemma 2.8. *Assume that*

$$\alpha > \gamma \qquad \text{and} \qquad \alpha \geq 1-\gamma, \tag{2.33}$$

and

$$\int d\nu_0 =: C_0 < \infty. \tag{2.34}$$

Then $\int d\nu_t$ is decreasing for all $t > 0$.

Proof. For the proof we use an equivalence between size distributions ν_t and so-called size orderings, which is also useful in the study of uniqueness of solutions (cf. [20], [22]).

For that, we introduce the cumulative distribution

$$\varphi(\lambda) := \int_{\lambda_-}^{\infty} d\nu_t,$$

which is decreasing and left continuous at jumps. Next we take the inverse of φ, such that we regard λ as a function of φ. The precise definition is

$$\lambda(t, x) := \sup\{y \mid \varphi(t, y) > x\},$$

such that $\lambda(t, \cdot)$ is decreasing and right continuous. If ν_t is a weak solution of (1.34), (1.35), then λ satisfies

$$\begin{aligned} \partial_t \lambda(t, \varphi) &= \lambda^\alpha(t, \varphi) u(t) - \lambda^{\alpha-\gamma}(t, \varphi), \quad \text{for a.e. } t, \quad \text{if } \lambda(t, \varphi) > 0, \\ \int_0^\infty \lambda(t, \varphi)\, d\varphi &= \int_0^\infty \lambda_0(\varphi)\, d\varphi, \end{aligned}$$

and vice versa. The equivalence of these concepts of solutions has been established in [20], [22].

We notice that

$$\int d\nu_t = \sup\{\varphi \mid \lambda(t, \varphi) > 0\} =: \bar{\varphi}(t).$$

Hence, our aim is to show that if $\bar{\varphi}(0) < \infty$, then it is decreasing in time.

For the proof, we first recall that due to $\alpha \geq 1 - \gamma$ and $u \in L^2_{loc}([0, \infty))$, we have

$$\int \lambda^\alpha(t, \varphi)\, d\varphi \in L^\infty((0, \infty)) \qquad \text{and} \qquad \int \lambda^{\alpha-\gamma}(t, \varphi)\, d\varphi \in L^2_{\text{loc}}([0, \infty)).$$

Let ψ^η be a standard Dirac sequence. We extend $u(t)$ by zero to $(-\infty, 0)$ and let $u^\eta := u * \psi^\eta$, such that u^η is bounded uniformly in time in any finite interval.

We now define λ_η as the solution of

$$\begin{aligned} \partial_t \lambda_\eta &= \lambda_\eta^\alpha u^\eta(t) - \lambda_\eta^{\alpha-\gamma}, \quad \text{as long as } \lambda_\eta(t, \varphi) > 0, \\ \lambda_\eta(0, \varphi) &= \lambda_0(\varphi). \end{aligned} \tag{2.35}$$

With given bounded u^η, a solution is easily constructed by solving the corresponding differential equation in each point φ. Again, since u^η is bounded, once $\lambda(t_\varphi, \varphi) = 0$ it remains zero for all $t > t_\varphi$. Hence $\bar{\varphi}_\eta := \sup\{\varphi \mid \lambda_\eta(t, \varphi) > 0\}$ is decreasing in time.

We now argue that if $\lambda(t, \varphi)$ denotes the size ordering corresponding to ν_t, then

$$\int_0^\infty |\lambda_\eta(t, \varphi) - \lambda(t, \varphi)|\, d\varphi \to 0, \quad \text{as } \eta \to 0 \ \text{ for all } t > 0, \tag{2.36}$$

if (2.33) is satisfied.

Assume for the moment that (2.36) holds, which immediately implies for almost all t that $\bar{\varphi}(t) \leq \bar{\varphi}_\eta(t) \leq \bar{\varphi}(0) = C_0$. Then

$$\begin{aligned} u(t) &= \frac{\int \lambda^{\alpha-\gamma}(t, \varphi)\, d\varphi}{\int \lambda^\alpha(t, \varphi)\, d\varphi} \\ &\leq \frac{(\int \lambda(t, \varphi)\, d\varphi)^{\alpha-\gamma} (\bar{\varphi}(t))^{1-\alpha-\gamma}}{\int \lambda^\alpha(t, \varphi)\, d\varphi} \\ &\leq \frac{C C_0^{1-\alpha-\gamma}}{\int \lambda^\alpha(t, \varphi)\, d\varphi}. \end{aligned}$$

However, since $\int \lambda(t,\varphi)\,d\varphi \equiv const.$ for all $t \geq 0$, we find that for $t \leq T$ it must hold that $\int \lambda^{\alpha}(t,\varphi)\,d\varphi \geq c_T > 0$. Hence, for any finite time interval, u is bounded. But then again, as in the argument for $\bar{\varphi}_\eta$, $\bar{\varphi}(t) = \int d\nu_t$ is indeed decreasing for all $t > 0$.

To prove (2.36) we first notice that $\lambda_\eta - \lambda$ satisfies

$$\begin{aligned}\partial_t(\lambda_\eta - \lambda) &= u^\eta(\lambda_\eta^\alpha - \lambda^\alpha) - (\lambda_\eta^{\alpha-\gamma} - \lambda^{\alpha-\gamma}) + \lambda^\alpha(u^\eta - u)\\ &=: f(\tilde{\lambda})(\lambda_\eta - \lambda) + \lambda^\alpha(u^\eta - u), \end{aligned} \tag{2.37}$$

for some $\tilde{\lambda}(t,\varphi) \in (\lambda_\eta(t,\varphi), \lambda(t,\varphi))$ with

$$f(\lambda) := \alpha\lambda^{\alpha-1}u^\eta - (\alpha-\gamma)\lambda^{\alpha-\gamma-1}.$$

We find by elementary calculus that f attains its maximum at

$$\lambda_0 = \left(\frac{1}{u^\eta}\frac{(\alpha-\gamma)(\alpha-\gamma-1)}{\alpha(\alpha-1)}\right)^{1/\gamma} =: (u^\eta)^{-1/\gamma}c_{\alpha,\gamma},$$

and due to $\alpha > \gamma$ we find $\lambda_0 > 0$. Hence, this implies

$$f(\lambda_0) \leq C_{\alpha,\gamma}(u^\eta)^{1-(\alpha-1)/\gamma} \leq C_{\alpha,\gamma}(u^\eta)^2, \tag{2.38}$$

where the last inequality follows if $\alpha \geq 1-\gamma$. Since $u \in L^2_{loc}([0,\infty))$, we have $(\lambda_\eta - \lambda)(\cdot,\varphi) \in H^1_{loc}([0,\infty))$ for all φ, and thus it follows that $\partial_t(\lambda_\eta - \lambda)\mathrm{sign}(\lambda_\eta - \lambda) = \partial_t|\lambda_\eta - \lambda|$ (cf. e.g. [12], Lemma 7.6). Now, we multiply (2.37) by $\mathrm{sign}(\lambda_\eta - \lambda)$ and integrate to obtain that

$$\begin{aligned}\int_0^\infty |\lambda_\eta(t,\varphi) - \lambda(t,\varphi)|\,d\varphi &\leq \int_0^t |u^\eta - u| \int_0^\infty \lambda^\alpha(s,\varphi)\,d\varphi\\ &\qquad \int_0^t\int_0^\infty f(\tilde{\lambda})|\lambda_\eta(s,\varphi) - \lambda(s,\varphi)|\,d\varphi\,ds\end{aligned}$$

and (2.38) implies

$$\begin{aligned}\int_0^\infty |\lambda_\eta(t,\varphi) - \lambda(t,\varphi)|\,d\varphi &\leq \int_0^t |u^\eta - u| \int_0^\infty \lambda^\alpha(s,\varphi)\,d\varphi\\ &\quad + C\int_0^t |u^\eta|^2 \int_0^\infty |\lambda_\eta(s,\varphi) - \lambda(s,\varphi)|\,d\varphi\,ds.\end{aligned}$$

Now $\int \lambda^\alpha(t,\varphi)\,d\varphi \in L^\infty((0,T))$, $u^\eta \to u$ in $L^2((0,T))$ and u^η is uniformly bounded in $L^2((0,T))$ for any $T < \infty$. Hence we obtain (2.36) via Gronwall's lemma. □

2.4. A Sequence of Data Mimicking Nucleation

In this section we construct a sequence of data which is motivated by numerical simulations of the large-time behavior of the Becker-Döring equations in [5].

A striking similarity is observed between the following two scenarios. First, one computes for given density ρ and finite system size n the equilibrium solution $c^n = (c_l^n)$, given by

$$c_l^n = Q_l z_n^l, \quad l \le n, \qquad z_n \quad \text{such that} \sum_{l=1}^{n} l c_l^n = \rho. \tag{2.39}$$

Clearly, for given $\rho > \rho_s$ and n there exists $z_n > z_s$ such that (2.39) is satisfied. Furthermore, as $n \to \infty$, it must hold that $z_n \to z_s$, since otherwise (2.39) could not be satisfied.

If one plots z_n versus n (cf. Fig. 2.2 in [5]), one observes that z_n decreases rapidly for small n before it reaches a plateau where the rate of change with n is extremely small. The width of this plateau rapidly increases as $\rho - \rho_s$ decreases.

In the simulations for the Becker-Döring equations with data $c_1(0) = \rho$, one obtains the same picture: First $c_1(t)$ decreases rapidly, then it reaches a plateau, which corresponds to the metastable state (with the same value as the plateau for z_n), before it finally converges steadily to equilibrium (cf. Fig. 4.1 in [5]).

These observations suggest that in order to mimic nucleation of large clusters, i.e., the way the system leaves the metastable state, one might consider the sequence of equilibria for finite system size n, and replace $t \to \infty$ by $n \to \infty$.

More precisely, we define for given $\varepsilon > 0$ a number $n = n(\varepsilon)$ and $c^n = (c_l^n)$ such that

$$c_l^n = \begin{cases} Q_l z_n^l : & l = 1, \dots, n \\ 0 : & l \ge n \end{cases},$$

with z_n such that (2.39) is satisfied and n is such that

$$\begin{aligned} F(c^{n-1}) &:= \sum_{l=1}^{n-1} c_l^{n-1} \left(\ln \left(\frac{c_l^{n-1}}{c_l^s} \right) - 1 \right) + c_l^s \\ &\ge \varepsilon^\gamma \\ &> \sum_{l=1}^{n} c_l^n \left(\ln \left(\frac{c_l^n}{c_l^s} \right) - 1 \right) + c_l^s = F(c^n). \end{aligned} \tag{2.40}$$

Indeed, this is possible, since $F(c^n) \to 0$ as $n \to \infty$, which follows from the continuity of F under weak$*$ convergence, which has been established in [2]. For the convenience of the reader, we show here that $F(c^n) \to 0$. For that we recall

$$F(c^n) = \sum_{l=1}^{n} c_l^s f \left(\frac{c_l^n - c_l^s}{c_l^s} \right),$$

with $f(z) = (1+z)\ln(1+z) - z$. Since $f(z) \le z^2$ and due to (1.8), we find for $m < \infty$ that

$$\sum_{l=1}^{m} c_l^s f \left(\frac{c_l^n - c_l^s}{c_l^s} \right) \le \sum_{l=1}^{m} \frac{|c_l^n - c_l^s|^2}{c_l^s} \le C e^{m^{1-\gamma}} \sum_{l=1}^{m} |c_l^n - c_l^s|^2, \tag{2.41}$$

which for fixed m converges to zero, since $z_n \to z_s$.

On the other hand,

$$\sum_{l=m}^{n} c_l^n \left(\ln \left(\frac{c_l^n}{c_l^s} \right) - 1 \right) + c_l^s = \sum_{l=m}^{n} c_l^n \ln \frac{1}{c_l^s} + \sum_{l=m}^{n} c_l^n (\ln c_l^n - 1) + c_l^s, \tag{2.42}$$

and with (1.8),

$$\sum_{l=m}^{n} c_l^n \ln \frac{1}{c_l^s} = \frac{q}{z_s(1-\gamma)} \sum_{l=m}^{n} l^{1-\gamma} c_l^n + o\left(\sum_{l=m}^{n} l^{1-\gamma} c_l^n \right). \tag{2.43}$$

Furthermore, for small $\eta > 0$,

$$\sum_{l=m}^{n} c_l^n (\ln c_l^n - 1) \le C \left(\sum_{l=m}^{n} |c_l^n|^{1-\eta} + \sum_{l=m}^{n} |c_l^n|^{1+\eta} \right)$$

and

$$\sum_{l=m}^{n} |c_l^n|^{1+\eta} \le \rho^\eta \sum_{l=m}^{n} |c_l^n| \le \frac{\rho^{1+\eta}}{m}. \tag{2.44}$$

In addition we estimate

$$\begin{aligned} \sum_{l=m}^{n} |c_l^n|^{1-\eta} &\le \sum_{l=m}^{n} l^{2\eta} l^{-2\eta} |c_l^n|^{1-\eta} \\ &\le \left(\sum_{l=m}^{n} l^{2\eta/(1-\eta)} |c_l^n| \right)^{1-\eta} \left(\sum_{l=m}^{n} l^{-2} \right)^{\eta} \\ &\le \rho^{1-\eta} m^{3\eta-1} m^{-\eta} = \rho^{1-\eta} m^{2\eta-1}. \end{aligned} \tag{2.45}$$

Finally, as we have seen now several times,

$$\sum_{l=m}^{n} c_l^s \le C e^{-m^{1-\gamma}}. \tag{2.46}$$

Thus, summarizing (2.41)–(2.46), we find

$$\begin{aligned} F(c^n) &\le C e^{m^{1-\gamma}} \sum_{l=1}^{m} |c_l^n - c_l^s|^2 + C \left(\sum_{l=m}^{n} l^{1-\gamma} c_l^n + m^{-1+\eta} \right) \\ &\le C e^{m^{1-\gamma}} \sum_{l=1}^{m} |c_l^n - c_l^s|^2 + C(m^{-\gamma} + m^{-1+\eta}). \end{aligned}$$

Hence, we can first choose m large and then n large enough to conclude that $F(c^n) \to 0$ as $n \to \infty$.

Lemma 2.9. *If (2.40) and (2.39) are satisfied, then the following inequalities hold:*

$$\frac{1}{C}\varepsilon^\gamma \;\le\; (z_{n-1}-z_s) \le C\varepsilon^\gamma, \tag{2.47}$$

$$\frac{1}{C\varepsilon} \;\le\; n \le \frac{C}{\varepsilon}, \tag{2.48}$$

$$\sum_{l=l_0}^{n} c_l^n \;\le\; C\varepsilon, \tag{2.49}$$

with l_0 *as in (1.30), i.e.,* $l_0 = \varepsilon^{-x}$ *for some* $x \in (0, 1/2)$.

Proof. We first notice that due to $c_l^{n-1} > c_l^s$ we have

$$\begin{aligned}\varepsilon^\gamma \;\le F(c^{n-1}) \;&=\; \sum_l^{n-1} c_l^{n-1}\ln\left(\frac{c_l^{n-1}}{c_l^s}\right) - \sum_l^{n-1}(c_l^{n-1}-c_l^s)\\ &\le\; \ln\left(\frac{z_{n-1}}{z_s}\right)\sum_l^{n-1} l Q_l z_{n-1}^l\\ &=\; \ln\left(\frac{z_{n-1}}{z_s}\right)\rho \le \frac{(z_{n-1}-z_s)}{z_s}\rho,\end{aligned} \tag{2.50}$$

which proves the first inequality in (2.47).

Now we consider (2.39), i.e.,

$$\rho = \sum_{l=1}^{n-1} l c_l^{n-1} = \sum_{l=1}^{n-1} l Q_l z_{n-1}^l \approx z_s \sum_{l=1}^{n-1} l \exp\left\{-\frac{q}{z_s(1-\gamma)} l^{1-\gamma} + l \ln\frac{z_{n-1}}{z_s}\right\}. \tag{2.51}$$

We recall that, for large l due to (1.8),

$$\ln(l Q_l z_{n-1}^l) \sim l\ln\left(\frac{z_{n-1}}{z_s}\right) - \frac{q}{z_s(1-\gamma)} l^{1-\gamma} + O(l^{1-2\gamma}).$$

Hence, (2.51) can only be satisfied if

$$(n-1)\ln\left(\frac{z_{n-1}}{z_s}\right) \le C(n-1)^{1-\gamma} \tag{2.52}$$

holds uniformly in n, and and with (2.50) we find

$$n-1 \le \frac{C}{\varepsilon} \qquad \text{and} \qquad n \le \frac{C+1}{\varepsilon}. \tag{2.53}$$

To prove the lower bound on n, we first recall Lemma 2.1 which implies

$$\sum_{l=1}^{n} l^{1-\gamma}(c_l^n - c_l^s) \;\le\; \varepsilon^{\gamma/2}, \tag{2.54}$$

$$\sum_{l=l_0}^{n} l^{1-\gamma}(c_l^n - c_l^s) \;\le\; \varepsilon^{\gamma}. \tag{2.55}$$

First, we notice that (2.54) implies

$$\varepsilon^{\gamma/2} \geq \frac{1}{n^\gamma} \sum_{l=1}^{n} l(c_l^n - c_l^s) = \frac{1}{n^\gamma}(\rho - \rho_s),$$

such that

$$n \geq \frac{1}{C\varepsilon^{1/2}}, \tag{2.56}$$

and in particular $n \gg l_0$.

We now claim that for sufficiently small δ, one finds

$$\sum_{l=\delta n}^{n} l(c_l^n - c_l^s) \geq \frac{\rho - \rho_s}{2}. \tag{2.57}$$

First, we recall

$$\sum_{l=\delta n}^{n} l(c_l^n - c_l^s) \;=\; \rho - \rho_s + \sum_{l=n}^{\infty} l c_l^s - \sum_{l=l_0}^{\delta n} l(c_l^n - c_l^s) - \sum_{l=1}^{l_0} l(c_l^n - c_l^s). \tag{2.58}$$

With (2.53) and (2.56) we have

$$\sum_{l=n}^{\infty} l c_l^s \leq n^{1+\gamma} e^{-c_0 n^{1-\gamma}} \ll \varepsilon.$$

Furthermore, using (2.54), we find

$$\sum_{1}^{l_0} l(c_l^n - c_l^s) \leq C l_0^\gamma \varepsilon^{\gamma/2} \leq C\varepsilon^{\gamma(1/2-x)}.$$

Finally, (2.53) implies

$$\sum_{l_0}^{\delta n} l(c_l^n - c_l^s) \leq \delta n^\gamma \varepsilon^\gamma \leq C\delta,$$

and thus (2.57) follows from (2.58).

Now (2.57) implies together with (2.55) that

$$\varepsilon^\gamma \geq \sum_{\delta n}^{n} l^{1-\gamma}(c_l^n - c_l^s) \geq \frac{1}{n^\gamma} \sum_{\delta n}^{n} l(c_l^n - c_l^s) \geq \frac{\rho - \rho_s}{2n^\gamma},$$

and hence

$$n \geq \frac{1}{C\varepsilon}. \tag{2.59}$$

Then, the upper bound on $z_{n-1} - z_s$ follows from (2.52) and (2.59).

Finally, to prove (2.49), we first notice that due to (2.59) we find for some $\delta > 0$ that

$$\sum_{\delta n}^{n} (c_l^n - c_l^s) \leq \frac{\rho - \rho_s}{\delta n} \leq \frac{C}{\delta} \varepsilon.$$

On the other hand, for a suitable $c_0 > 0$, one has

$$\sum_{l_0}^{\delta n} (c_l^n - c_l^s) \leq \sum_{l_0}^{\delta n} e^{l \ln\left(\frac{z_n}{z_s}\right) - c_0 l^{1-\gamma}}.$$

For sufficiently small δ we have

$$l \ln\left(\frac{z_n}{z_s}\right) - c_0 l^{1-\gamma} \leq -\frac{c_0}{2} l^{1-\gamma},$$

for $l \in (l_0, \delta n)$. Hence

$$\sum_{l_0}^{\delta n} (c_l^n - c_l^s) \leq C l_0^{\gamma} e^{-\frac{c_0}{2} l_0^{1-\gamma}} \leq C\varepsilon,$$

if l_0 satisfies (1.30). This finishes the proof of (2.49) and hence the proof of the lemma. □

We remark that for the data constructed in this section the rescaled densities ν_0^{ε} have uniformly compact support due to (2.48). However, even though the numerical simulations suggest that this sequence of data resembles closely the average behavior of the solutions, as well as the behavior of the small clusters, to conclude also on the detailed behavior of the tail might be a bit daring.

3. Higher-Order Dynamics

In this section we go a step further than in Section 2 and provide a detailed asymptotic expansion of solutions to the Becker-Döring equation in the parameter ε. In the first part we compute the expansion of the equation, whereas the second part provides an expansion of the energy identity of the Becker-Döring equations, which gives additional energy-type identities on the different levels of the expansion.

3.1. Expansion of the Equation

In the following we describe in detail how to systematically derive an asymptotic expansion for the solution of the Becker-Döring equations in the large-time regime, i.e., for the solution of

$$\partial_t c_l = \frac{1}{\varepsilon^{1-\alpha+\gamma}} (J_{l-1} - J_l), \quad l \geq 2, \tag{3.1}$$

where c_1 is determined by

$$\sum_{l=1}^{\infty} l c_l(t) = \rho, \quad \text{for all } t \geq 0. \tag{3.2}$$

We choose the following ansatz for the expansion for the large and small clusters respectively for some powers $\{x_i\}_i$, $\{y_i\}_i$, which have to be determined. In the following, l_0 will denote as before the cut between small and large clusters and can be chosen to be $l_0 \sim C|\ln \varepsilon|$ or $l_0 \sim \varepsilon^{-x}$, $x \in (0, 1/2)$, for example.

Large cluster expansion:

$$c_l(t) = \varepsilon^2 \left(\nu_0(t, \lambda) + \sum_{i=1}^{\infty} \varepsilon^{x_i} \nu_i(t, \lambda) \right), \quad l \geq l_0, \ \lambda = \varepsilon l, \tag{3.3}$$

Small cluster expansion:

$$c_l - c_l^s = \varepsilon^\gamma \left(f_{0,l} + \sum_{i=1}^{\infty} \varepsilon^{y_i} f_{i,l} \right), \quad 2 \leq l, \tag{3.4}$$

where $f_{i,l}$ denotes the l-th component of a sequence f_i.

Since the monomers play a special role, we use the following notation.

Monomer expansion:

$$c_1 - z_s = \varepsilon^\gamma \left(u_0 + \sum_{i=1}^{\infty} \varepsilon^{y_i} u_i \right). \tag{3.5}$$

The scale ε^2 for the large clusters and ε^γ for the small clusters has been justified heuristically in Section 1.4 of the introduction and rigorously in Section 2; hence, we do not repeat the argument here.

For $l \geq l_0$ we write

$$\begin{aligned} J_l &= (a_l(c_1 - z_s) - q/l^\gamma)c_l - (b_{l+1}c_{l+1} - b_l c_l) \\ &\approx \varepsilon^{2-\alpha+\gamma}[(\lambda^\alpha(u_0 + \cdots) - q\lambda^{\alpha-\gamma})(\nu_0 + \cdots) \\ &\quad - \varepsilon^{1-\gamma}\partial_\lambda(z_s \lambda^\alpha \nu_0 + \cdots)] \end{aligned}$$

and find with (3.1) that

$$\begin{aligned} 0 = \; & \partial_t \left(\nu_0 + \sum_{i=1}^{\infty} \varepsilon^{x_i} \nu_i \right) \\ & + \partial_\lambda \left(\lambda^\alpha \left(\varepsilon^y \left(u_0 + \sum_{i=1}^{\infty} \varepsilon^{y_i} u_i \right) - \frac{q\varepsilon^\gamma}{\lambda^\gamma} \right) \left(\nu_0 + \sum_{i=1}^{\infty} \varepsilon^{x_i} \nu_i \right) \right. \\ & \left. - \varepsilon^{1-\gamma} \partial_\lambda \left(\lambda^\alpha \left(z_s + \frac{\varepsilon^\gamma q}{\lambda^\gamma} \right) \left(\nu_0 + \sum_{i=1}^{\infty} \varepsilon^{x_i} \nu_i \right) \right) \right), \end{aligned} \tag{3.6}$$

whereas the constraint (3.2) gives

$$\begin{aligned}\rho - \rho_s &= \sum_{l=l_0}^{\infty} l c_l - \sum_{l=l_0}^{\infty} l c_l^s + \sum_{l=1}^{l_0-1} l(c_l - c_l^s) \\ &= \int \lambda \nu_0 \, d\lambda + \sum_{i=1}^{\infty} \varepsilon^{x_i} \int \lambda \nu_i \, d\lambda \\ &\quad + \varepsilon^{\gamma} \left(\sum_{l=1}^{l_0-1} l f_{0,l} + \sum_{i=1}^{\infty} \varepsilon^{y_i} \sum_{l=1}^{l_0-1} l f_{i,l} \right) + o(\varepsilon). \qquad (3.7)\end{aligned}$$

Collecting the leading order terms gives the now well-known

$$\begin{aligned}\partial_t \nu_0 + \partial_\lambda \left(\lambda^\alpha \left(u_0 - \frac{q}{\lambda} \right) \nu_0 \right) &= 0, \\ \int \lambda \nu_0 \, d\lambda &= \rho - \rho_s, \qquad (3.8)\end{aligned}$$

which implies

$$u_0 = \frac{q \int \lambda^{\alpha-\gamma} \, \nu_0 \, d\lambda}{\int \lambda^\alpha \, \nu_0 \, d\lambda}. \qquad (3.9)$$

As we have seen before, to leading order, the evolution of the large clusters decouples from the evolution of the small clusters.

To compute higher-order expansions, we now turn to the small clusters. First, recall that $a_l z_s c_l^s = b_{l+1} c_{l+1}^s$ such that we can rewrite the flux as

$$\begin{aligned}J_l &= a_l c_1 c_l - b_{l+1} c_{l+1} \\ &= a_l z_s (c_l - c_l^s) - b_{l+1}(c_{l+1} - c_{l+1}^s) + a_l c_l^s (c_1 - z_s) \\ &\quad + a_l (c_1 - z_s)(c_l - c_l^s), \qquad (3.10)\end{aligned}$$

and with (3.4) we can write

$$J_l = \varepsilon^\gamma \left(J_{0,l} + \sum_{i=1}^{\infty} \varepsilon^{y_i} J_{i,l} \right)$$

with

$$J_{0,l} = a_l z_s f_{0,l} - b_{l+1} f_{0,l+1} + a_l c_l^s u_0 \qquad (3.11)$$

and

$$J_{1,l} = a_l z_s f_{1,l} - b_{l+1} f_{1,l+1} + a_l c_l^s u_1 + a_l u_0 f_{0,l}, \qquad (3.12)$$

etc. Now, (3.1) implies

$$\begin{aligned}0 &= \varepsilon^{1-\alpha+\gamma} \partial_t \left(f_{0,l} + \sum_{i=1}^{\infty} \varepsilon^{y_i} f_{i,l} \right) \\ &\quad + J_{0,l-1} - J_{0,l} + \sum_{i=1}^{\infty} \varepsilon^{y_i} (J_{i,l-1} - J_{i,l}), \qquad (3.13)\end{aligned}$$

for all $l \geq 2$. Thus, we find

$$J_{0,l} \equiv const. =: C_0, \quad \text{for all } l.$$

With (3.11) and (3.4) this implies that $f_{0,l}$ is determined by

$$\begin{aligned} f_1^0 &= u_0, \\ f_{0,l+1} &= \frac{1}{b_{l+1}}(a_l z_s f_{0,l} + a_l c_l^s u_0 + C_0). \end{aligned} \tag{3.14}$$

We see that, if $C_0 \neq 0$, then $|f_{0,l}|$ grows very fast as $l \to \infty$, which would not agree with (3.7). Hence, we conclude $C_0 = 0$ and we easily derive by induction that

$$f_{0,l} = l Q_l z_s^{l-1} u_0 = l c_l^s \frac{u_0}{z_s}, \quad \text{for all } l \geq 1. \tag{3.15}$$

Hence, $f_{0,l}$ is in quasi-steady equilibrium with the monomer-density u_0, which is determined by the evolution of the large clusters. With (3.15) we compute

$$\lim_{l_0 \to \infty} \sum_{l=1}^{l_0-1} l f_{0,l} = \lim_{l_0 \to \infty} \frac{u_0}{z_s} \sum_{l=1}^{l_0-1} l^2 c_l^s = \frac{u_0}{z_s} \sum_{l=1}^{\infty} l^2 c_l^s =: \frac{u_0}{z_s} \Lambda_0, \tag{3.16}$$

where the constant Λ_0 only depends on the parameters in the system and can in principle be computed explicitly.

Plugging (3.16) into (3.7), we find

$$x_1 = \gamma$$

and

$$\int \lambda \nu_1 \, d\lambda = -\frac{u_0}{z_s} \Lambda_0. \tag{3.17}$$

Going back to (3.6) we see that a sensible expansion is possible if $n\gamma = 1$ for some $n = 2, 3, \ldots$. This is the case for all the examples given in Section 1.1, so we assume it here. In addition, whether or not the second-order derivative plays a role depends now on whether $\gamma < \frac{1}{2}$. In the first case we find

$$\partial_t \nu_1 + \partial_\lambda \left(\lambda^\alpha \left(u_0 - \frac{q}{\lambda^\gamma} \right) \nu_1 + u^1 \lambda^\alpha \nu_0 \right) = 0, \tag{3.18}$$

whereas if $\gamma = \frac{1}{2}$, as in two-dimensional cluster growth, (3.18) is replaced by

$$\partial_t \nu_1 + \partial_\lambda \left(\lambda^\alpha \left(u_0 - \frac{q}{\lambda^\gamma} \right) \nu_1 + u^1 \lambda^\alpha \nu_0 \right) - \partial_\lambda^2 (z_s \lambda^\alpha \nu_0) = 0. \tag{3.19}$$

In both cases (3.17) is formally equivalent to

$$\int \lambda^\alpha \left(u_0 - \frac{q}{\lambda^\gamma} \right) \nu_1 + u^1 \int \lambda^\alpha \nu_0 = -\frac{d}{dt} \frac{u_0}{z_s} \Lambda_0. \tag{3.20}$$

This equation determines u_1.

Given u_1, we go back to (3.10) and find that the term

$$a_l(c_1 - z_s)(c_l - c_l^s) = \varepsilon^{2\gamma} a_l u_0 f_{0,l} + o(\varepsilon^{2\gamma})$$

gives

$$y_1 = \gamma.$$

From (3.13) it follows, since $\alpha < 1$, that $J_{1,l} = const.$, but similarly as for $J_{0,l}$ we can conclude that $J_{1,l} = 0$ for all l. Then, $f_{1,l}$ is determined via

$$\begin{aligned} f_1^1 &= u_1, \\ f_{1,l+1} &= \frac{1}{b_{l+1}}(a_l z_s f_{1,l} + a_l c_l^s u_1 + a_l u_0 f_{0,l}) \\ &= \frac{1}{b_{l+1}}\left(a_l z_s f_{1,l} + a_l c_l^s u_1 + a_l \frac{c_l^s}{z_s} l (u_0)^2\right), \end{aligned} \tag{3.21}$$

and thus, by induction we see that $f_{1,l}$ is given by

$$f_{1,l} = c_l^s \left(l \frac{u_1}{z_s} + l(l-1)\left(\frac{u_0}{z_s}\right)^2 \right). \tag{3.22}$$

With this characterization of $f_{1,l}$ we can return to (3.7) to find $x_2 = \gamma$. We compute

$$\begin{aligned} \sum_{l=1}^{l_0-1} l f_{1,l} &= \frac{u_1}{z_s}\sum_{l=1}^{l_0-1} l^2 c_l^s + \left(\frac{u_0}{z_s}\right)^2 \sum_{l=1}^{l_0-1} l^2(l-1)c_l^s \\ &\to \frac{u_1}{z_s}\sum_{l=1}^{\infty} l^2 c_l^s + \left(\frac{u_0}{z_s}\right)^2 \sum_{l=1}^{\infty} l^2(l-1)c_l^s \\ &=: \frac{u_1}{z_s}\Lambda_0 + \left(\frac{u_0}{z_s}\right)^2 \Lambda_1, \end{aligned}$$

and the equation for ν_2 is given by by (3.6), which implies for $\gamma = 1/3$,

$$\partial_t \nu_2 + \partial_\lambda \left(\lambda^\alpha \left(u_0 - \frac{q}{\lambda^\gamma} \right) \nu_0 + \lambda^\alpha u_1 \nu^1 + \lambda^\alpha u_2 \nu_0 \right) = \partial_\lambda (z_s \lambda^\alpha \nu_0).$$

On the other hand, for $\gamma = 1/2$ we find

$$\partial_t \nu_2 + \partial_\lambda \left(\lambda^\alpha \left(u_0 - \frac{q}{\lambda^\gamma} \right) \nu_0 + \lambda^\alpha u_1 \nu^1 + \lambda^\alpha u_2 \nu_0 \right) = \partial_\lambda (q \lambda^{\alpha-\gamma} \nu_0 + z_s \lambda^\alpha \nu_1),$$

and u_2 has to be such that

$$\frac{u_1}{z_s}\Lambda_0 + \left(\frac{u_0}{z_s}\right)^2 \Lambda_1 = -\int \lambda \nu_2 \, d\lambda$$

is satisfied. We can summarize the general procedure:

Equation for large clusters + constraint	==>	(ν_0, u_0)
Equation for small clusters + u_0	==>	$(f_{0,l})_l$
Equation for large clusters + constraint + $f_{0,l}$	==>	(ν_1, u^1)
Equation for small clusters + u_1	==>	$(f_{1,l})_l$
...		
...		

Obviously we can continue this procedure to compute even higher-order expansions. We do not want to give the details here, but let us just mention that one obtains an expansion in ε^γ and we notice that α enters at some later stage through (3.13), and a useful expansion is possible if α is a multiple of γ (which is satisfied by the examples in Section 1.1).

3.2. Expansion of the Energy Estimate

In this section we show how a careful expansion of the energy estimate for the Becker-Döring equations enables one to identify further equalities in addition to (1.37).

For simplicity we confine ourselves in this section to the case of diffusion-controlled cluster growth in 3D, i.e., to the specific coefficients $\alpha = \gamma = 1/3$. We recall (2.1), i.e.,

$$F(c(t)) + \frac{1}{\varepsilon}\int_0^t D(c(s))\,ds = F(c(0)) = \varepsilon^{1/3}, \tag{3.23}$$

with

$$F(c(t)) = \sum_{l=1}^{\infty} c_l\left(\ln\left(\frac{c_l}{c_l^s}\right) - 1\right) + c_l^s = \sum_{l=1}^{\infty} c_l f\left(\frac{c_l - c_l^s}{c_l}\right),$$

where $f(z) = (1+z)\ln(1+z) - z$, and

$$D(c(t)) = \sum_{l=1}^{\infty} J_l \ln\left(\frac{a_l c_1 c_l}{b_{l+1}c_{l+1}}\right) \geq \sum_{l=1}^{\infty} \frac{|J_l|^2}{\max(a_l c_1 c_l, b_{l+1}c_{l+1})}. \tag{3.24}$$

We first consider the energy for large clusters

$$F_{\text{large}} := \sum_{l=l_0}^{\infty} c_l\left(\ln\left(\frac{c_l}{c_l^s}\right) - 1\right) + c_l^s.$$

For that we compute with (1.8) that

$$\begin{aligned}
\ln\left(\frac{1}{Q_l z_s^l}\right) &= \ln\left(\frac{a_l}{C_0 z_s}\right) + \sum_{k=2}^{l}\ln\left(1 + \frac{q}{z_s k^{1/3}}\right) \\
&\approx \ln\left(\frac{a_l}{C_0 z_s}\right) + \sum_{k=2}^{l}\left(\frac{q}{z_s k^{1/3}} - \frac{q^2}{z_s^2}\frac{1}{2k^{2/3}} + \cdots\right) \\
&\approx \ln\left(\frac{a_l}{C_0 z_s}\right) + \ln C + \frac{3q}{2z_s}l^{2/3} - \frac{3q^2}{2z_s^2}l^{1/3} + \cdots,
\end{aligned}$$

and hence

$$\sum_{l=l_0}^{\infty} c_l \ln\left(\frac{1}{Q_l z_s^l}\right) \approx \frac{3q}{2z_s}\sum_{l=l_0}^{\infty} l^{2/3} c_l - \frac{3q^2}{2z_s^2}\sum_{l=l_0}^{\infty} l^{1/3} c_l + o\left(\sum_{l=l_0}^{\infty} l^{1/3} c_l\right). \tag{3.25}$$

Furthermore, the remaining terms in F_{large} can be estimated similarly to those in Section 2.4 and thus shown to be of higher order:

$$\left|\sum_{l=l_0}^{\infty} c_l(\ln c_l - 1) + c_l^s\right| \le o\left(\sum_{l=l_0}^{\infty} l^{1/3} c_l\right).$$

Thus, (3.25) leads to

$$\begin{aligned}\varepsilon^{-1/3} F_{\mathrm{large}} \ = \ & \frac{3q}{2z_s}\int \lambda^{2/3}\,(\nu_0 + \varepsilon^{1/3}\nu_1 + \cdots) \\ & - \varepsilon^{1/3}\frac{3q^2}{2z_s^2}\int \lambda^{1/3}\nu_0 + \cdots. \end{aligned} \tag{3.26}$$

For the small clusters we recall

$$F_{\mathrm{small}} := \sum_{l=1}^{l_0-1} c_l\left(\ln\left(\frac{c_l}{c_l^s}\right) - 1\right) + c_l^s = \sum_{l=1}^{l_0-1} c_l^s f\left(\frac{c_l - c_l^s}{c_l^s}\right),$$

with $f(z) = (1+z)\ln(1+z) - z$. For small z, a good approximation should be $f(z) \approx \frac{1}{2}z^2$, and we write

$$F_{\mathrm{small}} = \tfrac{1}{2}\sum_{l=1}^{l_0-1}\frac{(c_l - c_l^s)^2}{c_l^s} = \varepsilon^{2/3}\tfrac{1}{2}\sum_{l=1}^{l_0-1}\frac{|f_{0,l}|^2}{c_l^s} + o(\varepsilon^{2/3}).$$

Recalling (3.15) and (3.16), we find

$$\varepsilon^{-2/3} F_{\mathrm{small}} = \frac{|u_0|^2}{2z_s^2}\sum_{l=1}^{l_0-1} l^2 c_l^s + o(1) = \frac{|u_0|^2}{2z_s^2}\Lambda_0 + o(1).$$

Next, we expand the energy dissipation rate.

First, we recall that due to $J_l = o(\varepsilon^{2/3})$ we have

$$\varepsilon^{-4/3}\sum_{l=1}^{l_0-1} J_l \ln\left(\frac{a_l c_1 c_l}{b_{l+1} c_{l+1}}\right) = o(\varepsilon^{1/3}).$$

To expand the dissipation rate for the large clusters, we will make the assumption that in view of (3.24) a good approximation is

$$\begin{aligned}\varepsilon^{-4/3} D_{\mathrm{large}} \ := \ & \varepsilon^{-4/3}\sum_{l=l_0}^{\infty} J_l \ln\left(\frac{a_l c_1 c_l}{b_{l+1} c_{l+1}}\right) \\ \approx \ & \frac{1}{2\varepsilon^{4/3}}\sum_{l=l_0}^{\infty}\frac{|J_l|^2}{a_l c_1 c_l} + \frac{1}{2\varepsilon^{4/3}}\sum_{l=l_0}^{\infty}\frac{|J_l|^2}{b_{l+1} c_{l+1}} \\ \approx \ & \tfrac{1}{2}\int \frac{|v^\varepsilon|^2}{\lambda^{1/3} c_1 \nu} + \tfrac{1}{2}\int \frac{|v^\varepsilon|^2}{\lambda^{1/3}(z_s + \frac{q\varepsilon^{1/3}}{\lambda^{1/3}})\nu}, \end{aligned} \tag{3.27}$$

with

$$v^{\varepsilon} = (\lambda^{1/3}u_0 - q)\nu_0 + \varepsilon^{1/3}[(\lambda^{1/3}u_0 - q)\nu_1 + u^1\lambda^{1/3}\nu_0] + o(\varepsilon^{1/3}). \tag{3.28}$$

We expand the denominators

$$\begin{aligned} \frac{1}{\lambda^{1/3}c_1\nu} &= \frac{1}{\lambda^{1/3}(z_s + \varepsilon^{1/3}u_0 + \cdots)(\nu_0 + \varepsilon^{1/3}\nu_1 + \cdots)} \\ &= \frac{1}{\lambda^{1/3}z_s\nu_0}\left(1 - \varepsilon^{1/3}\left(\frac{u_0}{z_s} + \frac{\nu_1}{\nu_0}\right) + o(\varepsilon^{1/3})\right) \end{aligned} \tag{3.29}$$

and

$$\frac{1}{\lambda^{1/3}(z_s + \frac{q\varepsilon^{1/3}}{\lambda^{1/3}})\nu} = \frac{1}{\lambda^{1/3}z_s\nu_0}\left(1 - \varepsilon^{1/3}\left(\frac{q}{z_s\lambda^{1/3}} + \frac{\nu_1}{\nu_0}\right) + o(\varepsilon^{1/3})\right). \tag{3.30}$$

Thus, collecting (3.27)–(3.30), we find

$$\begin{aligned} \varepsilon^{-4/3}D_{\text{large}} &= \int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_0 \\ &\quad + 2\varepsilon^{1/3}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_1 + 2\varepsilon^{1/3}\int(\lambda^{1/3}u_0 - q)\nu_0 \\ &\quad - \varepsilon^{1/3}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_1 - \frac{\varepsilon^{1/3}}{2}\frac{q}{z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{2/3}z_s}\nu_0 \\ &\quad - \frac{\varepsilon^{1/3}}{2}\frac{u_0}{z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_0 + o(\varepsilon^{1/3}). \end{aligned} \tag{3.31}$$

However, since

$$\int(\lambda^{1/3}u_0 - q)\nu_0 = 0,$$

equation (3.31) reduces to

$$\begin{aligned} \varepsilon^{-4/3}D_{\text{large}} &= \int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_0 \\ &\quad + \varepsilon^{1/3}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_1 \\ &\quad - \frac{\varepsilon^{1/3}}{2}\frac{q}{z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{2/3}z_s}\nu_0 \\ &\quad - \frac{\varepsilon^{1/3}}{2}\frac{u_0}{z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\nu_0 + o(\varepsilon^{1/3}). \end{aligned} \tag{3.32}$$

Comparing now all the terms in the energy estimate (3.23) and requiring that the terms with equal ε-powers add up to zero, we find first the well-known energy estimate for ν_0:

$$\frac{d}{dt}\left(\frac{3q}{2z_s}\int \lambda^{2/3}\,\nu_0\right) + \int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\,\nu_0 = 0.$$

The terms with power $\varepsilon^{1/3}$ give

$$\frac{d}{dt}\left(\frac{|u_0|^2}{2z_s^2}\Lambda_0 + \frac{3q}{2z_s}\int \lambda^{2/3}\,\nu_1 - \frac{3q^2}{2z_s^2}\int \lambda^{1/3}\,\nu_0\right) + \int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\,\nu_1 - \frac{q}{2z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{2/3}z_s}\,\nu_0 - \frac{u_0}{2z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\,\nu_0 = 0.$$

However, we notice that the terms with ν_0 do not involve any higher-order terms, and we expect that they should add up to zero. Indeed, one easily checks that

$$\frac{d}{dt}\left(\frac{3q^2}{z_s^2}\int \lambda^{1/3}\,\nu_0\right) + \frac{q}{z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{2/3}z_s}\,\nu_0 + \frac{u_0}{z_s}\int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\,\nu_0 = 0 \quad (3.33)$$

as well as

$$\frac{d}{dt}\left(\frac{|u_0|^2}{2z_s^2}\Lambda_0 + \frac{3q}{2z_s}\int \lambda^{2/3}\,\nu_1\right) + \int \frac{(\lambda^{1/3}u_0 - q)^2}{\lambda^{1/3}z_s}\,\nu_1 = 0, \quad (3.34)$$

which is just (3.35) for $\gamma = 1/3$.

It seems that (3.33) has in this form not been realized before, and we hope that it will also be helpful in a study of the large-time behavior of ν_0.

We conclude with the remark that the analogue of (3.34) for general coefficients satisfying $1 - 2\gamma > 0$ is given by

$$\frac{d}{dt}\left(\Lambda_0\frac{(u_0)^2}{2z_s^2} + \frac{q}{1-\gamma}\int \lambda^{1-\gamma}\,\nu_1\,d\lambda\right) + q\int \frac{(\lambda^{\alpha}u_0 - \lambda^{\alpha-\gamma})^2}{\lambda^{\alpha}}\,\nu_1\,d\lambda = 0. \quad (3.35)$$

A. Appendix

A.1. Derivation of Coefficients

In this appendix we briefly describe the standard heuristic derivation of coefficients for the Becker-Döring equations for diffusion-controlled and interface-reaction-controlled cluster growth in a first-order phase transition. We assume that the clusters are compact, that is, they have spherical shape, such that in three dimensions the radius r_l of the cluster is related to l by $\frac{4}{3}\pi r_l^3 = l$.

To compute the rate at which a monomer is attached to an l-cluster, one solves the steady-state diffusion equation for the local monomer density u with a sink boundary condition at the cluster, i.e., $u(r_l) = 0$, and the condition that, far away from the cluster, it is equal to the overall monomer density, i.e., $u(\infty) = c_1$. We compute that $u(r) = c_1(1 - r_l/r)$. The rate at which monomers hit the cluster is then given by $-4\pi D r_l^2 u'(r_l) = 4\pi D r_l c_1$ with the diffusivity constant D, which implies

$$a_l = 4\pi D r_l.$$

The rate at which monomers leave an l-cluster is computed similarly, assuming there is a sink of monomers at infinity and the density at the cluster boundary is in local equilbrium. The corresponding value is given by the Gibbs-Thomson formula as $z_s(1+\Gamma/r_l)$, where z_s is the density of monomers in equilibrium with a plane surface and Γ is proportional to surface tension. The solution to the quasi-steady diffusion field can again be computed and we arrive at

$$b_l = 4\pi D z_s(r_l+\Gamma) = a_l(z_s + q/l^{1/3}),$$

with $q = (\frac{4\pi}{3})^{1/3} z_s\Gamma$.

For two-dimensional systems one proceeds analogously to obtain

$$a_l = 2\pi D \qquad \text{and} \qquad b_l = 2\pi D z_s(1+\Gamma/r_l) = a_l(z_s + q/l^{1/2}),$$

with $q = \pi^{1/2} z_s \Gamma$.

In interface-reaction-controlled systems, the rate of change of the cluster size is assumed to be proportional to the surface of the interface times the difference between monomer density inside and outside the cluster. In three dimensions that means that the rate of change of cluster size is given by

$$4\pi r_l^2 k(c_1 - \bar{c}),$$

where k is a kinetic constant and $\bar{c}$ the concentration of monomers in the cluster. Again, if there is a sink of monomers, i.e., $\bar{c}=0$, then the rate at which monomers are attached is

$$a_l = 4\pi k r_l^2 = k(4\pi)^{2/3} 3^{1/3} l^{2/3},$$

whereas if there is a sink of monomers outside the cluster, and local equilibrium inside the cluster, then

$$b_l = 4\pi k r_l^2 z_s \left(1 + \frac{\Gamma}{r_l}\right) = a_l \left(z_s + \frac{(4\pi/3)^{1/3} z_s \Gamma}{l^{1/3}}\right).$$

Analogously one obtains in two dimensions

$$a_l = 2\pi k r_l = 2\pi^{1/2} k l^{1/2}$$

and

$$b_l = 2\pi k r_l z_s \left(1 + \frac{\Gamma}{r_l}\right) = a_l \left(z_s + \frac{\pi^{1/2} z_s \Gamma}{l^{1/2}}\right).$$

Acknowledgment

I am grateful to Philippe Laurençot for helpful comments on an early draft of Section 2.

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