

## THE BEKER–DÖRING SYSTEM AND ITS LIFSHITZ–SLYOZOV LIMIT\*

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**Abstract.** We investigate the connection between two classical models for the study of phase transition phenomena, the Becker–Döring equations, and the Lifshitz–Slyozov system. More precisely, we introduce a scaling parameter and show that the solution to the Becker–Döring equations converges to that of the Lifshitz–Slyozov system as the scaling parameter goes to 0.

**Key words.** Becker–Döring system, Lifshitz–Slyozov system, coagulation-fragmentation, phase transition

**AMS subject classifications.** 45K05, 82C26, 82C70, 35Q99

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**1. Introduction.** This paper addresses the mathematical connection between the infinite set of ordinary differential equations

$$(1.1) \quad \frac{d}{dt}c_i = J_{i-1} - J_i \quad \text{for } i \geq 2,$$

$$(1.2) \quad J_i = a_i c_1 c_i - b_{i+1} c_{i+1} \quad \text{for } i \geq 1,$$

$$(1.3) \quad \frac{d}{dt}c_1 = -2J_1 - \sum_{i=2}^{\infty} J_i,$$

$$(1.4) \quad c_i(t=0) = c_i^0 \quad \text{for } i \geq 1$$

and the integro-differential system

$$(1.5) \quad \partial_t f + \partial_x J = 0 \quad \text{for } t \geq 0, x \geq 0,$$

$$(1.6) \quad J(x, t) = (a(x)c(t) - b(x))f(t, x),$$

$$(1.7) \quad c(t) + \int_0^{\infty} x f(t, x) dx = \rho,$$

$$f(t=0, x) = f^0(x) \quad \text{for } x \geq 0,$$

$$(1.8) \quad c(t=0) = c^0, \quad \rho = c^0 + \int_0^{\infty} x f^0(x) dx.$$

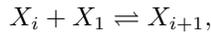
These two models are well known in the literature on phase transition phenomena; however, for the reader's convenience, let us briefly recall their physical background. In the Becker–Döring system (1.1)–(1.3), introduced in [4], the unknown is the infinite sequence of functions  $\{c_i, i \in \mathbb{N}^*\}$ , while  $a_i, b_i$  are given positive coefficients. The quantity  $c_i(t)$  for any  $i \geq 2$  represents the concentration of clusters containing  $i$ -particles (or  $i$ -mers, denoted by  $X_i$  in what follows) at time  $t$ , and  $c_1(t)$  denotes the concentration of free particles (or monomers, denoted by  $X_1$  in what follows).

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Intuitively,  $c_i$  may be thought of as the number of  $i$ -mers present in the solution. More precisely, if one used a finite-mass description of the system, denoting by  $n_i$  the total number of  $i$ -mers, then  $c_i(t)$  would be the expected value of this random variable at time  $t$ . Equation (1.1) is the kinetic equation for the  $c_i$ 's. It expresses the net rate of change of  $c_i$  due to the reactions



by which  $i$ -mers can gain (coagulation) monomers to become  $i+1$ -mers or, conversely, (fragmentation). The quantity  $J_i$  denotes the rate of this reaction. Its expression in terms of the concentrations is obtained from a straightforward application of mass-action kinetics, denoting by  $a_i$  and  $b_{i+1}$  the coagulation and fragmentation rates. Equation (1.3) expresses the net rate of change of  $c_1$ . (Note that monomers are involved in infinitely many reactions.) It can be shown (see [1]) to imply the following relation:

$$m_1 \sum_{i=1}^{\infty} i c_i(t) = \rho,$$

which means conservation of the total amount of monomers present in the solution. Here  $m_1$  denotes the mass of one monomer. The system is supplemented by the Cauchy data (1.4).

In the Lifshitz–Slyozov model (1.5)–(1.7), on the other hand, the size of clusters is thought of as a continuously varying variable  $x > 0$ , which will now replace  $i$ . The quantity  $f(t, x)$  designates the density of aggregates with respect to this size variable, and  $c(t)$  denotes the concentration of monomers. Equation (1.5) is the continuity equation in size space, whereas the third equation again represents the conservation of monomers. To obtain a precise expression of the flux  $J$ , one needs to derive an expression for the growth rate of grains, and this in turn requires a physical description of the interaction between clusters and monomers. By making assumptions on this mass-transfer mechanism (e.g., assuming it to be diffusion-limited, or surface-attached, or other), one may then obtain an explicit expression for the rate coefficients  $a$  and  $b$ . Details can be found in the original paper [14] or in the recent review [20]. Here we will restrict ourselves only to a more phenomenological level, in which the details of the cluster/monomer interaction are subsumed into the (God-given) coefficients  $a_i, b_i$  for the discrete case, or  $a(x), b(x)$  for the continuous case. Since these two models are obtained from rather different approaches, it seems interesting to investigate how one may be obtained from another. Since the Lifshitz–Slyozov model treats size as a continuous variable, it seems that it should be obtainable from the Becker–Döring one as either a “macroscopic” or “large time” limit. More precisely, since these models describe a later stage of coarsening, in which the average cluster size increases monotonically with time, it is physically reasonable that the large time dynamics of the Becker–Döring system should in a sense be close to that of the Lifshitz–Slyozov system. A precise mathematical formulation of this idea may be found in [19]. Here we will take a “macroscopic limit” point of view, much in the spirit (although the mathematical structure is simpler here) of hydrodynamic limits in fluid mechanics; see [10]. The mathematical theory of both systems is by now fairly complete. An extensive study of the Becker–Döring system for nonnegative initial data with finite total mass  $\rho$  may be found in [1]. In particular, the existence of solutions is proved for coefficients  $a_i, b_i = \mathcal{O}(i)$  (see Theorem 2.2 of [1]), whereas existence fails if the coefficients grow faster than  $i$ . Uniqueness is obtained under supplementary

assumptions on higher moments of the initial data, and a thorough description of the asymptotic behavior is given. Here we will place ourselves in situations where the existence and uniqueness results of [1] apply.

The well-posedness theory for the Lifshitz–Slyozov equations is discussed in [5] for smooth coefficients and in [12] for more general ones. Global existence and uniqueness is also discussed in [17]. The papers [16], [18] have very complete results on the asymptotic behavior, and [15] has a derivation of (1.5)–(1.7) via homogenization methods. A variant of this system in which encounters between clusters are taken into account is studied in [6] and [13].

The outline of the paper is as follows. In section 2, we introduce the relevant nondimensional numbers and rewrite the system (1.1)–(1.3) in dimensionless form. Sections 3 and 4 are devoted to the hyperbolic and parabolic scalings, respectively.

**2. Notation and dimensionless equations.** For a fixed value of the rescaling parameter  $\epsilon$ , we will use the results in [1] to obtain a uniquely defined solution (the precise definition of which may be found in [1]) to the rescaled problem as well as some estimates on its moments. More precisely, we will make use of the following results (see, respectively, Theorems 3.7 and 2.5 in [1]).

**THEOREM 2.1** (see [1]). *Assume that the initial data is nonnegative and satisfies  $\sum_{i=1}^{\infty} i c_i^0 < \infty$ . Assume that the coefficients satisfy  $a_i, b_i = \mathcal{O}(\sqrt{i})$ . Then the problem (1.1)–(1.4) admits one and only one solution.*

**THEOREM 2.2** (see [1]). *Let  $c$  be a solution to (1.1)–(1.3), and let  $\varphi_i$  be a non-negative sequence satisfying the following assumptions:*

$$(2.1) \quad \left\{ \begin{array}{l} \int_{t_1}^{t_2} \sum_{i=1}^{\infty} |\varphi_{i+1} - \varphi_i| a_i c_i(t) dt < \infty, \quad \sup_t \sum_{i=1}^{\infty} \varphi_i c_i(t) < \infty, \\ \varphi_{i+1} - \varphi_i \geq 0 \quad \text{for } i \text{ large enough.} \end{array} \right.$$

Then for any  $m \geq 2$  the following relation holds true:

$$(2.2) \quad \begin{aligned} \sum_{i=m}^{\infty} c_i(t_2) \varphi_i &+ \int_{t_1}^{t_2} \left\{ \sum_{i=m}^{\infty} b_{i+1} c_{i+1}(s) (\varphi_{i+1} - \varphi_i) \right\} ds \\ &= \sum_{i=m}^{\infty} c_i(t_1) \varphi_i + \int_{t_1}^{t_2} \left\{ \sum_{i=m}^{\infty} a_i c_1(s) c_i(s) (\varphi_{i+1} - \varphi_i) \right\} ds \\ &\quad + \int_{t_1}^{t_2} \varphi_m (a_{m-1} c_1(s) c_{m-1}(s) - b_m c_m(s)) ds. \end{aligned}$$

In the case where the kinetic coefficients are bounded, it follows from Theorem 2.2 in [1] that assumption (2.1) is satisfied for  $\varphi_i = 1$ , and  $\varphi_i = i^\alpha$  for any  $\alpha \geq 1$ . This will make it possible to derive estimates on the moments of the solution. We will also use (2.2) with sequences having only finitely many nonzero terms.

Before rescaling the system (1.1)–(1.3) in terms of a small parameter, we first rewrite it in dimensionless form. Let us summarize all of the absolute constants and reference quantities which will be used in what follows:

- $T$ : characteristic time,
- $C_1$ : characteristic value for the monomer concentration  $c_1$ ,
- $C$ : characteristic value for the large cluster concentration  $c_i, i \geq 2$ ,
- $A_1$ : characteristic value for the first coagulation coefficient  $a_1$ ,
- $A$ : characteristic value for the coagulation coefficients  $a_i, i \geq 2$ ,
- $B$ : characteristic value for the fragmentation coefficients  $b_i, i \geq 2$ ,

- $M$ : characteristic value for the total mass,
- $m_1$ : mass of one monomer,
- $\mathcal{N}$ : Avogadro number,
- $M_m$ : molar mass of monomers:  $M_m = \mathcal{N}m_1$ .

Note that due to the particular role played by monomers we will have  $A_1 \neq A$ , which means that the kinetic equation for  $c_2$ , which involves the quantity  $c_1^2$ , will be rescaled in a different way from the generic  $i$ th equation.

The corresponding dimensionless quantities are then defined as

$$(2.3) \quad \bar{t} = \frac{t}{T}, \quad \bar{c}_1(\bar{t}) = c_1(\bar{t}T)/C_1, \quad \bar{a}_1 = \frac{a_1}{A_1}, \quad \bar{\rho} = \frac{\rho}{M},$$

$$(2.4) \quad \bar{c}_i(\bar{t}) = c_i(\bar{t}T)/C, \quad \bar{a}_i = \frac{a_i}{A}, \quad \bar{b}_i = \frac{b_i}{B} \text{ for } i \geq 2.$$

When one rewrites (1.1)–(1.3) in terms of the new variables, the following dimensionless parameters appear:

$$(2.5) \quad \begin{cases} \gamma = C/C_1, & \mu = \frac{M\mathcal{N}}{M_m C_1} = \frac{M}{m_1 C_1}, \\ \alpha = ATC_1, & \alpha_1 = A_1 C_1/AC, \quad \beta = BT. \end{cases}$$

Omitting the overlines, the system takes the following dimensionless form:

$$(2.6) \quad \frac{d}{dt}c_i = \alpha(a_{i-1}c_1c_{i-1} - a_i c_1 c_i) + \beta(b_{i+1}c_{i+1} - b_i c_i) \quad \text{for } i > 2,$$

$$(2.7) \quad \frac{d}{dt}c_2 = \alpha_1 \alpha a_1 c_1^2 - \alpha a_2 c_1 c_2 + \beta(b_3 c_3 - b_2 c_2),$$

$$(2.8) \quad \frac{dc_1}{dt} = -\gamma \left[ 2(\alpha_1 \alpha a_1 c_1^2 - \beta b_2 c_2) + \sum_{i=2}^{\infty} (\alpha a_i c_1 c_i - \beta b_{i+1} c_{i+1}) \right].$$

As we noted above, by taking  $\varphi = i$  in (2.2), we obtain mass conservation:

$$(2.9) \quad c_1 + \gamma \sum_{i=2}^{\infty} i c_i = \mu \rho.$$

The dimensionless parameters defined in (2.5) appear as coefficients in this problem and need to be scaled appropriately with respect to  $\epsilon$ . Let us begin by explaining the heuristics of this choice. If the right-hand side in (2.6) is to yield a derivative, then  $\alpha$  and  $\beta$  should scale as the reciprocal of  $\epsilon$ . The quantity expressed in (2.9) is the total mass of the system. If this is to remain finite (and nonzero), then  $\mu$  should be of order 1. We would then like to view the sum over  $i$  in this equation as a Riemann sum for the integral in (1.7). Observing the system on a larger scale means taking  $x = \epsilon i$ ; thus  $\gamma$  should be taken of the same order as  $x dx$ , that is, the square of  $\epsilon$ .

More precisely, we are interested in the asymptotic behavior of large clusters whose size is on the order of  $1/\epsilon$ . For instance, if  $i$ -particles are spheres with radius  $L\bar{r}_i$ , where  $L$  is a characteristic length and  $\bar{r}_i$  is dimensionless, we have the relation  $L\bar{r}_i = (\frac{3V_m}{4\pi\mathcal{N}})^{1/3} i^{1/3}$ , which involves the molar volume of free atoms  $V_m$ . Hence we consider a situation in which  $1/\epsilon = \frac{4\pi L^3 \mathcal{N}}{3V_m}$  is large. The total mass of the aggregates (excluding monomers) is equal to  $m_1 C \sum_{i=2}^{\infty} i \bar{c}_i$ . Assuming that the average size is on the order of  $1/\epsilon$  and that all  $\bar{c}_i$ 's are of order 1, we see that the total mass is on

the order of  $C m_1/\epsilon^2$ . For that mass to be on the same order as the total mass of free monomers  $m_1 C_1$ , as well as the total mass, we therefore need to take  $\gamma = \epsilon^2$  and  $\mu = 1$  ( $C = \epsilon^2 C_1$ ,  $C_1 = M/m_1$ ). In the same way, we assume that for large clusters the fragmentation is of the same order as coagulation, and  $M$  is chosen accordingly:  $AC_1 = B$  ( $M = Bm_1/A$  and  $\alpha = \beta$ ). Clearly, if one phenomenon dominates the other, then in the continuous limit  $\epsilon \rightarrow 0$  one term vanishes, thus making the limiting regime either purely a coagulation regime or a fragmentation regime. In order to obtain nontrivial dynamics, it remains to fix the time scale with  $T = 1/(\epsilon B)$ , which means we study the large time behavior of the system.

These heuristic remarks naturally lead us to the following choice of the parameters in terms of  $\epsilon$ :

$$\gamma = \epsilon^2, \quad \mu = 1, \quad \alpha = \beta = 1/\epsilon, \quad \alpha_1 \leq 1.$$

At this point, a word of explanation about the bound imposed on  $\alpha_1$  is in order. This bound roughly says that, as the observational scale increases, the first reaction (encounter of two monomers to form a dimer) plays a negligible role. From the mathematical point of view, this means that we get rid of the negative power of  $\epsilon$  which this reaction introduces; from the physical point of view, it means that, in the macroscopic limit, we eliminate the coupling between (macroscopic) clusters and (microscopic) monomers introduced by this reaction. This was remarked upon in [7] and is reminiscent of the so-called infrared difficulty encountered when going from a granular to a continuous description of matter.

With this choice of the parameters, the rescaled version of (2.6)–(2.8) is

$$\begin{cases} \frac{d}{dt} c_i = \frac{1}{\epsilon} (a_{i-1} c_{i-1} - a_i c_i) c_1 + \frac{1}{\epsilon} (b_{i+1} c_{i+1} - b_i c_i) & \text{for } i > 2, \\ \frac{d}{dt} c_2 = \frac{1}{\epsilon} \alpha_1 a_1 c_1^2 - \frac{1}{\epsilon} a_2 c_1 c_2 + \frac{1}{\epsilon} (b_3 c_3 - b_2 c_2), \\ \frac{d}{dt} c_1 = -2(\epsilon a_1 \alpha_1 c_1^2 - \epsilon b_2 c_2) - \epsilon \sum_{i=2}^{\infty} (a_i c_i c_1 - b_{i+1} c_{i+1}). \end{cases}$$

This rescaled system is the starting point of our analysis. Throughout the remainder of the paper, we will denote by  $(c_i^\epsilon)_{i \geq 1}$  the solution to that system. The summation by parts formula corresponding to (2.2) is now (taking  $m = 2, t_1 = 0, t_2 = t$ )

$$\begin{aligned} \sum_{i=2}^{\infty} c_i^\epsilon(t) \varphi_i &+ \int_0^t \left\{ \sum_{i=2}^{\infty} \frac{b_{i+1}}{\epsilon} c_{i+1}^\epsilon(s) (\varphi_{i+1} - \varphi_i) \right\} ds \\ (2.10) \quad &= \sum_{i=2}^{\infty} c_i^\epsilon(0) \varphi_i + \int_0^t \left\{ \sum_{i=2}^{\infty} \frac{a_i}{\epsilon} c_1^\epsilon(s) c_i^\epsilon(s) (\varphi_{i+1} - \varphi_i) \right\} ds \\ &+ \int_0^t \varphi_2 \left( a_1 \frac{\alpha_1}{\epsilon} (c_1^\epsilon(s))^2 - \frac{b_2}{\epsilon} c_2^\epsilon(s) \right) ds. \end{aligned}$$

As mentioned above, in the case of bounded coefficients  $a_i$ , this formula holds true for  $\varphi_i = 1$  and  $\varphi_i = i^\alpha$  ( $\alpha \geq 1$ ). The choice  $\varphi_i = 1$  gives an estimate on the total number of clusters, whereas the choice  $\varphi_i = i$  yields mass conservation.

We will denote by  $f^\epsilon$  the function defined by

$$f^\epsilon(t, x) = c_i^\epsilon(t) \quad \text{if } t > 0, x_i = i\epsilon \leq x < x_{i+1} = (i+1)\epsilon \quad \text{for some } i \geq 2,$$

and  $f^\epsilon(t, x) = 0$  if  $x \in [0, 2\epsilon[$ . This choice of a piecewise constant function, with “mesh size”  $\epsilon$ , is convenient because sums such as the one in (2.9) will be handled as Riemann sums and will converge to the corresponding integral once it is known that the sequence  $(f^\epsilon)_{\epsilon>0}$  has a limit.

Our main result then shows that for  $\epsilon$  close to zero, the couple  $(c_1^\epsilon, f^\epsilon)$  is an approximate solution to the Lifshitz–Slyozov system.

**THEOREM 2.3.** *Assume the kinetic coefficients  $a_i, b_i$  satisfy*

$$(2.11) \quad a_i, b_i \leq K, \quad |a_{i+1} - a_i| \leq K/i, \quad |b_{i+1} - b_i| \leq K/i$$

for some constant  $K$ . Consider a sequence  $\epsilon_n \rightarrow 0$ .

Then there exist a subsequence, which we will still denote by  $\epsilon_n$ , and two functions  $a, b \in W_{loc}^{1,\infty}((0, \infty)) \cap L^\infty((0, \infty))$  such that, for any  $R > r > 0$ ,

$$(2.12) \quad \lim_{\epsilon_n \rightarrow 0} \sup_{i \in (r/\epsilon_n, R/\epsilon_n)} (|a_i - a(\epsilon_n i)| + |b_i - b(\epsilon_n i)|) = 0.$$

Assume, moreover, that there exist four constants  $0 < s \leq 1, M_0 < \infty, \rho < \infty, M_s < \infty$  for which, for all  $\epsilon > 0$ ,

$$(2.13) \quad \epsilon \sum_{i=2}^{\infty} c_i^{0,\epsilon} \leq M_0, \quad c_1^{0,\epsilon} + \epsilon^2 \sum_{i=2}^{\infty} i c_i^{0,\epsilon} = \rho, \quad \epsilon \sum_{i=2}^{\infty} (\epsilon i)^{1+s} c_i^{0,\epsilon} \leq M_s.$$

Then the subsequence  $\epsilon_n$  may be chosen in such a way that

$$\begin{cases} f^{\epsilon_n} \rightharpoonup f, \quad x f^{\epsilon_n} \rightharpoonup x f \text{ in } C^0([0, T]; \mathcal{M}^1(0, \infty) - \text{weak} - *), \\ c_1^{\epsilon_n}(t) \rightarrow c(t) \text{ uniformly in } C^0([0, T]), \end{cases}$$

where  $(c, f)$  is a solution to (1.5)–(1.8).

**REMARK 2.4.** *In our scaling approach, it is obvious that the coefficients  $a_i, b_i$  should not depend on  $\epsilon$ . Thus, for the continuous limits  $a, b$  to be locally bounded and Lipschitz (a very natural condition) and for convergence (2.12) to hold true, it appears that the boundedness condition (2.11) is in fact necessary. This means that condition (2.11) is optimal. Another approach, more numerical-analytic than modelling in spirit, could be to a priori fix the coefficients  $a$  and  $b$  and to introduce an  $\epsilon$ -dependence for the discrete coefficients in order to impose a convergence. In this case, condition (2.11) may be relaxed in order to allow growth of the coefficients for large sizes.*

**REMARK 2.5.** *Here  $\mathcal{M}^1(0, \infty)$  denotes the space of bounded measures on  $(0, \infty)$ , that is, the dual of the space  $C_0^0((0, \infty))$  of continuous functions vanishing at infinity and for  $x = 0$ ; see [8]. The map  $f$  in this context is a measure-valued solution to the Lifshitz–Slyozov equation (see [5] for a precise definition) so that, for any time  $t$ , we have  $f(t, \cdot) \in \mathcal{M}^1(0, \infty)$ .*

**REMARK 2.6.** *By condition (2.13), the initial data has to be prepared with respect to  $\epsilon$ . The condition on the zeroth moment means that the ratio of the initial number of agglomerates over  $C_1$  is of order  $\epsilon$  since we have*

$$C/C_1 \sum_{i=2}^{\infty} c_i^{0,\epsilon} = \epsilon^2 \sum_{i=2}^{\infty} c_i^{0,\epsilon} = \frac{\text{Initial number of agglomerates}}{C_1}.$$

**REMARK 2.7.** *This theorem gives useful information only when the system (1.5)–(1.8) is well-posed. This requires the characteristics of the transport equation (1.5)–(1.6) to point in the negative direction at  $x = 0$  (together with some regularity assumptions on the coefficients  $a$  and  $b$ ; we refer for detailed results to [5], [12]). This*

condition reads  $a(0)c(t) - b(0) \leq 0$ , which, for instance, follows from the stronger condition  $\rho \leq b(0)/a(0)$ . In this case, the uniqueness of the limit implies that the whole sequence converges in Theorem 2.3, provided (2.12) holds true. However, when this condition is not fulfilled, the system (1.5)–(1.8) has to be supplemented by a boundary condition at  $x = 0$ . We have not addressed this issue here, and finding the appropriate boundary condition is an open problem even from the physical point of view. A partial answer is given in section 4, in which we obtain a viscous correction to (1.5)–(1.8), together with a Dirichlet boundary condition, by making use of formal asymptotics. By extrapolating the result of [2] for scalar conservation laws (see also [3]), one might therefore conjecture that, in the case where  $c(t) \geq b(0)/a(0)$ , the correct boundary condition for (1.5)–(1.8) would be

$$(a(0)c(t) + b(0)) f(t, 0) = 2a_1c(t)^2.$$

**3. From Becker–Döring to Lifshitz–Slyozov.** Our theorem is merely a compactness statement about the sequence  $(c_1^\epsilon, f^\epsilon)$ . The compactness for  $f^\epsilon$  is obtained by showing boundedness of a higher moment, whereas the compactness for  $c_1^\epsilon$  is obtained by the use of the Arzelà–Ascoli theorem, in the spirit of [5]. These properties form the content of the next two lemmas.

LEMMA 3.1. *Assume that the kinetic coefficients verify (2.11) and that the initial data satisfy (2.13). Then for any  $T > 0$  there exists a constant  $C < \infty$  which depends only on  $s, M_0, M_s, \rho, K$ , and  $T$ , such that, for any  $\epsilon > 0$ ,*

$$\sup_{t \in [0, T]} \int_0^\infty (1 + x + x^{1+s}) f^\epsilon(t, x) dx \leq C.$$

*Proof.* The letter  $C$  will denote any constant depending on the data, as indicated in the statement of the result. Choosing  $\varphi_i = \epsilon$  in (2.10), we obtain

$$\begin{aligned} \int_0^\infty f^\epsilon(t, x) dx &= \epsilon \sum_{i=2}^\infty c_i^\epsilon(t) = \epsilon \sum_{i=2}^\infty c_i^{0, \epsilon} + \int_0^t [\alpha_1 a_1 (c_1^\epsilon(\tau))^2 - b_2 c_2^\epsilon(\tau)] d\tau \\ &\leq M_0 + \alpha_1 \rho^2 T. \end{aligned}$$

The estimate on the first moment may be derived similarly and is in fact derived in [1]; see (2.9). To deal with the higher moment, as a technical device we introduce the function  $e^\epsilon$  defined by

$$e^\epsilon(x) = x_i \quad \text{if } x_i = i\epsilon \leq x < x_{i+1} = (i + 1)\epsilon \quad \text{for some } i \geq 2,$$

and  $e^\epsilon(x) = 0$  if  $x \in [0, 2\epsilon[$ . We observe that for any positive  $r$  we have

$$\int_0^\infty (x/2)^r f^\epsilon(t, x) dx \leq \int_0^\infty (e^\epsilon(x))^r f^\epsilon(t, x) dx = \epsilon \sum_{i=2}^\infty (\epsilon i)^r c_i^\epsilon(t) \leq \int_0^\infty x^r f^\epsilon(t, x) dx.$$

We now take  $\varphi_i = \epsilon [e^\epsilon(i\epsilon)]^{1+s} = \epsilon [i\epsilon]^{1+s}$  in (2.10) to obtain

$$\begin{aligned} \int_0^\infty [e^\epsilon(x)]^{1+s} f^\epsilon(t, x) dx &= \epsilon \sum_{i=2}^\infty (\epsilon i)^{1+s} c_i^\epsilon(t) \\ &= \epsilon \sum_{i=2}^\infty (\epsilon i)^{1+s} c_i^{0, \epsilon} - \int_0^t \sum_{i=2}^\infty ((\epsilon i + \epsilon)^{1+s} - (\epsilon i)^{1+s}) \end{aligned}$$

$$\begin{aligned} & \cdot b_{i+1} c_{i+1}^\epsilon(\tau) d\tau \\ & + \int_0^t \sum_{i=2}^\infty ((\epsilon i + \epsilon)^{1+s} - (\epsilon i)^{1+s}) a_i c_i^\epsilon(\tau) c_1^\epsilon(\tau) d\tau \\ & + \int_0^t (2\epsilon)^{1+s} (\alpha_1 a_1 (c_1^\epsilon(\tau))^2 - b_2 c_2^\epsilon(\tau)) d\tau. \end{aligned}$$

In the right-hand side of this equality, the first term is less than  $M_s$ , and the fragmentation term has a negative contribution. (Fragmentation acts as a dissipation mechanism for moments of order larger than 1.) Using the fact that  $\epsilon < 1$  and  $s \leq 1$ , it is very easy to check that, for any  $i$ , we have

$$(\epsilon i + \epsilon)^{1+s} - (\epsilon i)^{1+s} \leq \epsilon(1 + s)(1 + \epsilon i).$$

We may therefore estimate the coagulation term as follows:

$$\begin{aligned} & \int_0^t \sum_{i=2}^\infty ((\epsilon i + \epsilon)^{1+s} - (\epsilon i)^{1+s}) a_i c_i^\epsilon(\tau) c_1^\epsilon(\tau) d\tau \\ & \leq K\rho \int_0^t \sum_{i=2}^\infty \epsilon c_i^\epsilon(\tau) (1 + s)(1 + \epsilon i) d\tau \\ & = K\rho (1 + s) \int_0^t \int_0^\infty (1 + e^\epsilon(x)) f^\epsilon(\tau, x) dx d\tau \\ & \leq K\rho (1 + s) \int_0^t \int_0^\infty (1 + x) f^\epsilon(\tau, x) dx d\tau \leq C, \end{aligned}$$

and this completes the proof of the lemma.  $\square$

LEMMA 3.2. *Assume that the kinetic coefficients verify (2.11) and that the initial data satisfy (2.13). Then, for any  $T > 0$ , the sequence of monomers concentrations  $(c_1^\epsilon)_{\epsilon > 0}$  is equicontinuous on  $[0, T]$ .*

*Proof.* By using mass conservation and formula (2.10), we can write

$$\begin{aligned} c_1^\epsilon(t+h) - c_1^\epsilon(t) &= -\epsilon^2 \sum_{i=2}^\infty i c_i^\epsilon(t+h) + \epsilon^2 \sum_{i=2}^\infty i c_i^\epsilon(t) \\ &= \epsilon \int_t^{t+h} \sum_{i=2}^\infty b_{i+1} c_{i+1}^\epsilon(\tau) d\tau - \epsilon \int_t^{t+h} \sum_{i=2}^\infty a_i c_i^\epsilon(\tau) c_1^\epsilon(\tau) d\tau \\ &\quad - \epsilon \int_t^{t+h} (2\alpha_1 a_1 (c_1^\epsilon(\tau))^2 - b_2 c_2^\epsilon(\tau)) d\tau \\ &= - \int_t^{t+h} \int_0^\infty (a^\epsilon(x) c_1^\epsilon(\tau) - b^\epsilon(x)) f^\epsilon(\tau, x) dx d\tau \\ &\quad - \epsilon \int_t^{t+h} (2\alpha_1 a_1 (c_1^\epsilon(\tau))^2 - b_2 c_2^\epsilon(\tau)) d\tau, \end{aligned}$$

where  $a^\epsilon, b^\epsilon$  are the piecewise constant functions defined from the  $a_i, b_i$ 's in the same way as  $f^\epsilon$  was defined from the  $c_i^\epsilon$ 's. On one hand, the space integral satisfies

$$\begin{aligned} \left| \int_t^{t+h} \int_0^\infty (a^\epsilon(x) c_1^\epsilon(\tau) - b^\epsilon(x)) f^\epsilon(\tau, x) dx d\tau \right| &\leq K(1 + \rho) \int_t^{t+h} \int_0^\infty f^\epsilon(\tau, x) dx d\tau \\ &\leq C(T)h \end{aligned}$$

by using Lemma 3.1. On the other hand, the boundary terms are estimated as follows:

$$0 \leq \int_t^{t+h} 2\epsilon\alpha_1 a_1(c_1^\epsilon(\tau))^2 d\tau \leq 2K\rho^2\epsilon h,$$

and

$$\begin{aligned} 0 \leq \int_t^{t+h} \epsilon b_2 f^\epsilon(\tau, 2\epsilon) d\tau &\leq K \int_t^{t+h} \epsilon \sum_{i=2}^\infty f^\epsilon(\tau, i\epsilon) d\tau \\ &\leq K \int_t^{t+h} \int_0^\infty f^\epsilon(\tau, x) dx d\tau \leq C(T)h, \end{aligned}$$

again by using the estimate of Lemma 3.1. Therefore, we conclude that  $|c_1^\epsilon(t+h) - c_1^\epsilon(t)|$  tends to 0 as  $h$  goes to 0, uniformly with respect to  $\epsilon > 0$  and  $t \in [0, T]$ .  $\square$

*Proof of Theorem 2.3.* By the Arzelà–Ascoli theorem and Lemma 3.2, we can suppose that

$$c_1^\epsilon(t) \rightarrow c(t) \text{ uniformly in } C^0([0, T]).$$

Let  $\varphi$  be a smooth test function supported in  $[\delta, R]$  for some  $0 < \delta < R < \infty$ . We set

$$\varphi^\epsilon(x) = \varphi(i\epsilon) \quad \text{for } i\epsilon \leq x < (i+1)\epsilon.$$

Since, for  $x > 0$ , we have

$$|\varphi(x) - \varphi^\epsilon(x)| \leq \epsilon \|\varphi'\|_{L^\infty},$$

$\varphi^\epsilon$  converges uniformly to  $\varphi$  on  $[0, \infty[$ . We now use (2.10) with  $\varphi_i = \epsilon\varphi(i\epsilon)$ . For  $\epsilon < \delta/2$ , the boundary term in (2.10) disappears by using the support property of  $\varphi$ . Therefore, we obtain

$$\begin{aligned} (3.1) \quad &\int_0^\infty f^\epsilon(t, x)\varphi^\epsilon(x) dx - \int_0^\infty f^\epsilon(0, x)\varphi^\epsilon(x) dx \\ &= \int_0^t \int_0^\infty f^\epsilon(\tau, x)(a^\epsilon(x)c_1^\epsilon(\tau)\Delta^+\varphi^\epsilon(x) + b^\epsilon(x)\Delta^-\varphi^\epsilon(x)) dx d\tau, \end{aligned}$$

where we have set

$$\Delta^\pm \varphi^\epsilon(x) = \frac{\varphi^\epsilon(x \pm \epsilon) - \varphi^\epsilon(x)}{\epsilon}.$$

It is straightforward to check that, as  $\epsilon \rightarrow 0$ , the quantity  $\Delta^\pm \varphi^\epsilon$  converges to  $\pm\varphi'$  uniformly on  $[0, \infty[$ .

Note that (3.1) and the estimates of Lemma 3.1 also prove that the sequence  $(\int_0^\infty f^\epsilon(t, x)\varphi^\epsilon(x) dx)_{\epsilon>0}$  is equicontinuous on  $[0, T]$  and equibounded. Therefore, by the Arzelà–Ascoli theorem, it belongs to a compact set of  $C^0([0, T])$ . Moreover, since  $\varphi^\epsilon$  approaches  $\varphi$  uniformly on  $\mathbb{R}^+$ , one deduces that  $(\int_0^\infty f^\epsilon(t, x)\varphi(x) dx)_{\epsilon>0}$  lies in a compact set of  $C^0([0, T])$ . By density, we realize that this also holds true in the case when  $\varphi \in C_0^0((0, \infty))$ . Now, by using the separability of  $C_0^0((0, \infty))$  and the Cantor diagonal process, we obtain for a subsequence (still denoted by  $\epsilon$  for simplicity) the following convergence for any  $\varphi \in C_0^0((0, \infty))$  and any  $T > 0$ :

$$\int_0^\infty \varphi f^\epsilon(t, x) dx \rightarrow \int_0^\infty \varphi f(t, dx)$$

as  $\epsilon$  goes to 0, uniformly on  $[0, T]$  for some  $f \in C^0([0, \infty); \mathcal{M}^1(0, \infty) - weak - *)$ . By virtue of the estimates of Lemma 3.1, we also know that

$$(1 + |x|^s)f \in L^\infty([0, \infty); \mathcal{M}^1(0, \infty)).$$

Therefore, we obtain the convergence results stated in the theorem. In particular, at  $t = 0$ , the limit  $f(t = 0, x)$  is given by the limit of  $f^{\epsilon_n}(0, x)$  constructed from the  $c_i^{0, \epsilon_n}$ 's.

Now we are able to pass to the limit in (3.1). The hypotheses on the kinetic coefficients ensure that, at least for a subsequence,  $a^\epsilon$  and  $b^\epsilon$  converge uniformly on any compact set  $[\delta, M]$  to some continuous functions  $a(x), b(x)$ . Let us postpone to the appendix the proof of this claim. Then, by using the compactness result of Lemma 3.2, we get

$$\begin{aligned} \int_0^\infty \varphi(x) f(t, dx) &= \int_0^\infty \varphi(x) f(0, dx) \\ &\quad + \int_0^t \int_0^\infty (a(x)c_1(\tau) - b(x))\varphi'(x) f(\tau, dx) d\tau, \end{aligned}$$

which corresponds to the transport equation (1.5). It remains to show that the conservation relation also passes to the limit. However, this follows easily from the uniform convergence  $e^\epsilon(x) \rightarrow x$  and the extra-moment estimate in Lemma 3.1 with  $s > 0$ ; we thus have

$$\lim_{\epsilon \rightarrow 0} \left( c_1^\epsilon(t) + \int_0^\infty e^\epsilon(x) f^\epsilon(t, x) dx \right) = \rho = c(t) + \int_0^\infty x f(t, dx).$$

Note that we are not able to determine the limit of  $\int_0^\infty f^\epsilon(t, x) dx$  because of the possible concentration of  $f^\epsilon$  as a Dirac mass at  $x = 0$ . We conclude that  $(c, f)$  is a solution to (1.5)–(1.7), and this completes the proof of the theorem.

**4. Second-order expansion: The modified Lifshitz–Slyozov model.** The connection between the discrete model (1.1)–(1.3) and the continuous one as the reference size becomes large may also give some insight into a physically relevant boundary condition for the Fokker–Planck-like equation presented in [9], [11]. For the sake of simplicity, let us take  $\alpha_1 = 1$ . Then (2.10) may be rewritten as follows:

$$\begin{aligned} &\sum_{i=2}^\infty c_i^\epsilon(t_2)\varphi_i - \sum_{i=2}^\infty c_i^\epsilon(t_1)\varphi_i \\ &= \int_{t_1}^{t_2} \left( \sum_{i=2}^\infty a_i c_1^\epsilon(\tau) c_i^\epsilon(\tau) \frac{\varphi_{i+1} - \varphi_{i-1}}{2\epsilon} + \sum_{i=2}^\infty a_i c_1^\epsilon(\tau) c_i^\epsilon(\tau) \frac{\varphi_{i+1} - 2\varphi_i + \varphi_{i-1}}{2\epsilon} \right) d\tau \\ &\quad + \int_{t_1}^{t_2} \left( - \sum_{i=2}^\infty b_i c_i^\epsilon(\tau) \frac{\varphi_{i+1} - \varphi_{i-1}}{2\epsilon} + \sum_{i=2}^\infty b_i c_i^\epsilon(\tau) \frac{\varphi_{i+1} - 2\varphi_i + \varphi_{i-1}}{2\epsilon} \right) d\tau \\ &\quad + \int_{t_1}^{t_2} \left[ \frac{\varphi_2}{\epsilon} a_1 (c_1^\epsilon(\tau))^2 - \frac{\varphi_1}{\epsilon} b_2 c_2^\epsilon(\tau) \right] d\tau. \end{aligned}$$

In order to keep the meaningful terms of order  $\epsilon$ , we slightly modify the interpolation. Omitting the exponent  $\epsilon$ , we define

$$(4.1) g(x) = c_i \text{ for } x \in [\epsilon(i - 3/2), \epsilon(i - 1/2)), \quad i \geq 2, \quad g(x) = 0 \text{ for } x \in [0, \epsilon/2),$$

with similar definitions for  $a$  and  $b$ . We choose  $\varphi_i = \varphi(\epsilon(i - 1))$ , where  $\varphi$  is a smooth  $\epsilon$ -independent function. Then the above equation can be rewritten as the following integral relation:

$$(4.2) \quad \begin{aligned} \frac{d}{dt} \int_0^\infty g(t, x)\varphi(x) dx &= \int_0^\infty (a(x)c(t) - b(x))g(t, x)\varphi'(x) dx \\ &+ \epsilon/2 \int_0^\infty (a(x)c(t) + b(x))g(t, x)\varphi''(x) dx \\ &+ \varphi(\epsilon)a_1c(t)^2 - \varphi(0)b_2c_2(t) + \mathcal{O}(\epsilon^2). \end{aligned}$$

The quantity  $c_2$  cannot be related to the trace at  $x = 0$  of the limit (as  $\epsilon \rightarrow 0$ ) of the function  $g$ . Therefore, we choose a test function which vanishes at the origin. The formal asymptotics is obtained by replacing the  $\mathcal{O}(\epsilon^2)$ -term by 0. Integrating by parts twice, we recover in this way the weak form of the following ( $\epsilon$ -dependent) Fokker–Planck equation

$$(4.3) \quad \begin{cases} \frac{\partial}{\partial t}g + \frac{\partial}{\partial x}G(g; t, x) = 0, \\ G(g; t, x) = (a(x)c(t) - b(x))g - \epsilon \frac{\partial}{\partial x} \left( \frac{a(x)c(t) + b(x)}{2} g(t, x) \right), \end{cases}$$

supplemented by the mass-conservation law

$$(4.4) \quad c(t) + \int_0^\infty xg(t, x) dx = \rho.$$

This model was proposed in [9], [11] as a variant of the Lifshitz–Slyozov model (1.5)–(1.7).

For (4.2) to be the weak form of (4.3), we need the boundary terms from either relation to be the same, which amounts to (as is easily obtained from one integration by parts)

$$\epsilon \left( \frac{a(0)c(t) + b(0)}{2} g(t, 0) \right) \varphi'(0) = \varphi(\epsilon)a_1c(t)^2.$$

Using the first-order approximation  $\varphi(\epsilon) = \epsilon\varphi'(0) + \mathcal{O}(\epsilon^2)$ , we obtain the following boundary condition:

$$(4.5) \quad (a(0)c(t) + b(0)) g(t, 0) = 2 a_1 c(t)^2.$$

Summarizing, we have obtained the following proposition.

**PROPOSITION 4.1.** *The function  $g$  defined by (4.1) and  $c$  is an approximate solution to the modified Lifshitz–Slyozov equation (4.3)–(4.4) with boundary condition (4.5) in the sense that, for any test function  $\varphi$  satisfying  $\varphi(0) = 0$ , we have*

$$\begin{aligned} \frac{d}{dt} \int_0^\infty g(t, x)\varphi(x) dx &= \int_0^\infty (a(x)c(t) - b(x))g(t, x)\varphi'(x) dx \\ &+ \epsilon/2 \int_0^\infty (a(x)c(t) + b(x))g(t, x)\varphi''(x) dx + \epsilon\varphi'(0)a_1c^2(t) + \mathcal{O}(\epsilon^2). \end{aligned}$$

As a final remark, we note that this boundary condition makes it possible to uniquely identify detailed balance equilibria. More precisely, a detailed balance equilibrium is a particular steady-state solution (we will denote it by  $m$  in analogy with

Maxwellian functions) for which the flux is identically zero:  $G(m) = 0$ . This ordinary differential equation may be readily integrated to obtain (see [9] for details)

$$m(x) = m(0) \frac{a(0)c + b(0)}{a(x)c + b(x)} \exp\left(\frac{2}{\epsilon} \int_0^x \frac{a(y)c - b(y)}{a(y)c + b(y)} dy\right).$$

Combining this relation and (4.5), we obtain

$$(4.6) \quad m(x) = c \frac{2 a_1 c}{a(x)c + b(x)} \exp\left(\frac{2}{\epsilon} \int_0^x \frac{a(y)c - b(y)}{a(y)c + b(y)} dy\right).$$

This explicit formula shows that the situation here is in complete analogy with the Becker–Döring system: the equilibrium is parametrized by the monomer concentration  $c$  and is not monodisperse (unlike what happens in the classical Lifshitz–Slyozov case).

Furthermore, for a given total mass  $\rho > 0$ , there exists a unique detailed balance equilibrium: indeed, the relation

$$\rho = c + \int_0^\infty xm(x) dx,$$

combined with (4.6), can be recast in the form of one equation  $F(c) = \rho$ . An easy computation then shows that the map  $F$  is strictly increasing. Finally, in the case where the function  $\frac{a}{b}$  has a finite limit for large sizes, one sees from (4.6) that  $F(c)$  will be finite only when  $c$  is less than a certain value, which has the physical meaning of a saturation concentration. Therefore, this diffusive model has a saturation threshold, in analogy with what occurs for the Becker–Döring system.

**Appendix. Uniform convergence of  $\mathbb{P}_0$  interpolants.**

LEMMA A.1. *Let  $u_i$  be a real sequence. Assume there exists  $K > 0$  such that, for all  $i \geq 0$ ,*

$$|u_i| \leq K, \quad |u_{i+1} - u_i| \leq K/i.$$

Define

$$u^\epsilon(x) = u_i \quad \text{for } i\epsilon = x_i \leq x < (i + 1)\epsilon = x_{i+1}.$$

Then, up to a subsequence,  $u^\epsilon$  converges uniformly on any interval  $[r, R]$ , with  $0 < r < R < \infty$ , to a continuous function  $u \in W^{1,\infty}([r, +\infty[)$ .

*Proof.* We shall show that  $u^\epsilon$  is  $\epsilon$ -close to a  $\mathbb{P}_1$ -sequence of functions verifying the requirements of the Arzelà–Ascoli theorem. Indeed, let us set

$$v^\epsilon(x) = \frac{u_{i+1} - u_i}{\epsilon} (x - \epsilon i) + u_i \text{ for } x_i \leq x < x_{i+1}.$$

On  $[x_i, x_{i+1}[$ , we get

$$|v^\epsilon(x) - u^\epsilon(x)| = \frac{|u_{i+1} - u_i|}{\epsilon} (x - \epsilon i) \leq \frac{K\epsilon}{i\epsilon} \leq \epsilon \frac{2K}{x},$$

which proves that

$$\sup_{x \geq r > 0} |v^\epsilon(x) - u^\epsilon(x)| \leq \epsilon \frac{2K}{r}.$$

On the other hand,  $v^\epsilon$  belongs to  $W^{1,\infty}$  with derivative satisfying on  $[x_i, x_{i+1}[$

$$\left| \frac{d}{dx} v^\epsilon(x) \right| = \frac{|u_{i+1} - u_i|}{\epsilon} \leq \frac{K}{i\epsilon} \leq \frac{2K}{x}.$$

Hence  $\frac{d}{dx} v^\epsilon(x)$  is bounded in  $L^\infty([r, +\infty[)$ . The result now follows from the Arzelà–Ascoli theorem.  $\square$

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