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QUASI STEADY STATE APPROXIMATION OF THE SMALL CLUSTERS IN BECKER-DÖRING EQUATIONS LEADS TO BOUNDARY CONDITIONS IN THE LIFSHITZ-SLYOZOV LIMIT

JULIEN DESCHAMPS[†], ERWAN HINGANT[‡], AND ROMAIN YVINEC[§]

Abstract. The following paper addresses the connection between two classical models of phase transition phenomena describing different stages of clusters growth. The first one, the Becker-Döring model (BD) that describes discrete-sized clusters through an infinite set of ordinary differential equations. The second one, the Lifshitz-Slyozov equation (LS) that is a transport partial differential equation on the continuous half-line $x \in (0, +\infty)$. We introduce a scaling parameter $\varepsilon > 0$, which accounts for the grid size of the state space in the BD model, and recover the LS model in the limit $\varepsilon \to 0$. The connection has been already proven in the context of outgoing characteristic at the boundary $x = 0$ for the LS model when small clusters tend to shrink. The main novelty of this work resides in a new estimate on the growth of small clusters, which behave at a fast time scale. Through a rigorous quasi steady state approximation, we derive boundary conditions for the incoming characteristic case, when small clusters tend to grow.

Key words. Becker-Döring system, Lifshitz-Slyozov equation, Boundary value for transport equation, quasi-steady state approximation, hydrodynamic limit.

AMS subject classifications. 34E13, 35F31, 82C26, 82C70

1. Introduction

This paper addresses the mathematical connection between two classical models of phase transition phenomena, describing different stages of the growth of clusters (or polymers, or aggregates). The first one is the Becker-Döring model (BD), first introduced in [3], which describes the early stages of cluster growth, at a small scale. Clusters are made of *elementary* particles and may increase or decrease their size, one-by-one, capturing (aggregation process) or shedding (fragmentation process) one particle, according to the set of chemical reactions:

$$C_1 + C_i \rightleftharpoons C_{i+1} \quad i \geq 1,$$

where C_i stands for a cluster of size i (consisting of i particles), while C_1 is a *free* elementary particle. In its mean-field version, the BD model is an infinite set of ordinary differential equations representing the time evolution of each concentration (number per unit of volume) of clusters made of i particles. In this work we focus on a dimensionless BD model that involves a small parameter $\varepsilon > 0$. We detail the standard scaling procedure in Appendix. Denote by $c_i^\varepsilon(t)$ the concentration at time $t \geq 0$ of clusters consisting of $i \geq 2$ particles and $u^\varepsilon(t)$ the concentration of free particles (clusters of size 1), where

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we make explicit the dependence on $\varepsilon>0$. The dimensionless system reads:

$$\begin{aligned} \frac{d}{dt}u^\varepsilon &= -\varepsilon J_1^\varepsilon - \varepsilon \sum_{i\geq 1} J_i^\varepsilon, \qquad t\geq 0, \\ \frac{d}{dt}c_i^\varepsilon &= \frac{1}{\varepsilon}\Big[J_{i-1}^\varepsilon - J_i^\varepsilon\Big], \quad i\geq 2,\ t\geq 0, \end{aligned} \tag{1.1}$$

where fluxes are defined by:

$$J_1^\varepsilon = \alpha^\varepsilon (u^\varepsilon)^2 - \varepsilon^\eta \beta^\varepsilon c_2^\varepsilon, \quad \text{and} \quad J_i^\varepsilon = a_i^\varepsilon u^\varepsilon c_i^\varepsilon - b_{i+1}^\varepsilon c_{i+1}^\varepsilon, \quad i\geq 2. \tag{1.2}$$

Here, coefficients a_i^ε and b_{i+1}^ε, for $i\geq 2$, denote respectively the rates of aggregation and fragmentation (ε-dependent), while α^ε and β^ε denote respectively the first rate of aggregation ($i=1$) and the first rate of fragmentation ($i=2$). Finally, η is an exponent standing for the strength of the first fragmentation rate, in which the results strongly depend (see also Section 7 for discussions). Observe that such model (at least formally) preserves the total number of particles (no source nor sink), that is

$$u^\varepsilon(t) + \sum_{i\geq 2} \varepsilon^2 i c_i^\varepsilon(t) = m^\varepsilon, \quad \forall t\geq 0. \tag{1.3}$$

The constant m^ε is entirely determined by the initial conditions at $t=0$ given by $u^{\text{in},\varepsilon}$ and $(c_i^{\text{in},\varepsilon})_{i\geq 2}$, non-negatives and ε-dependents. For theoretical studies on the well-posedness and long-time behavior of the deterministic Becker-Döring model (with $\varepsilon=1$), we refer the interested readers to [1,20,28,32] among many others.

The second model of phase transition, is the Lifshitz-Slyozov model (LS) introduced in [22]. This classically describes the late phase of cluster growth, at a "macroscopic scale". The LS model consists in a partial differential equation (of nonlinear transport type) representing the time evolution of the size distribution function $f(t,x)$ of clusters of (continuous) size $x>0$ at time $t\geq 0$, together with an equation stating the conservation of matter,

$$\begin{aligned} &\frac{\partial f}{\partial t} + \frac{\partial[(a(x)u(t)-b(x))f(t,x)]}{\partial x} = 0, \qquad t\geq 0,\ x>0, \\ &u(t) + \int_0^\infty x f(t,x) = m, \qquad\qquad t\geq 0, \end{aligned} \tag{1.4}$$

where a and b are functions of the size, respectively for the aggregation and fragmentation rates. The constant m plays the same role as in the BD model. Various authors studied this equation when the flux point outward at $x=0$ (*i.e.* when small clusters tend to fragment), namely if a condition like $a(0)u(t)-b(0)<0$ holds, see [10,18,19,24,25] among others for theoretical studies and technical assumptions. Indeed, in that case, uniqueness of weak solutions to the limit system (1.4) holds. But, recent applications in biology have raised the problem to include *nucleation* in the equation (*i.e.* small clusters tend to aggregate), for instance in [2,15,29]. These cases consider fluxes that point inward at $x=0$, thus the LS equation lacks a *boundary condition* to be well-posed. In particular, general coagulation-fragmentation (or nucleation-aggregation) model applied to amyloid fibrils formation are developed in [2,15], where Eq. (1.4), with incoming fluxes, appears as a building block of an integro-differential operator. Also, the authors

in [29] considered a pure aggregation model to fit Polyglutamine *in-vitro* polymerization experiments, with $b(x)=0$ and $a(0)>0$. It is common in the aforementioned literature to use a boundary condition of the type

$$a(0)u(t)f(t,0)=N(u(t)), \tag{1.5}$$

which couples the behavior of small clusters to the free particles' concentration $u(t)$. Such expression is typically justified by some pre-equilibrium hypothesis derived from a microscopic nucleation model. Zero-flux boundary condition is also traditionally imposed when one chooses to ignore nucleation, or if small clusters are assumed to instantaneously degrade into free particles (for instance when the first fragmentation rate is too strong), see for instance the prion equation [12, 14, 21]. Finally, let us mention that second-order expansion of the BD model yields a modified LS model for which a boundary condition is also needed. Some boundary condition was conjectured in this framework, *e.g.* in [8, 9, 11], but never rigorously proved.

In this work we aim to recover a solution of the LS equation and to construct proper boundary conditions, departing from the BD system Eq. (1.1) as the parameter ε goes to 0. This connection has been proved in [9, 20] for the classical case of outgoing characteristics. The authors have represented the dynamics of the BD model by a density function on a continuous size space. Accordingly, the size of each cluster is represented by a continuous variable $x>0$, and we let, for all $\varepsilon>0$,

$$f^{\varepsilon}(t,x)=\sum_{i\geq 2} c_i^{\varepsilon}(t)\mathbf{1}_{\Lambda_i^{\varepsilon}}(x), \quad x\geq 0,\ t\geq 0, \tag{1.6}$$

where for each $i\geq 2$, we defined $\Lambda_i^{\varepsilon}=[(i-1/2)\varepsilon,(i+1/2)\varepsilon)$. We denote for the remainder $f^{\mathrm{in},\varepsilon}:=f^{\varepsilon}(0,x)$. Hence, each cluster of (discrete) size initially $i\geq 2$ is seen as a cluster of size roughly $i\varepsilon\in\mathbb{R}_+$. The scaling used in the dimensionless BD equations (1.1) consists in an acceleration of the fluxes (by $1/\varepsilon$) thus a cluster can reach an *asymptotically infinite* size $i=x/\varepsilon$ in finite time. Then, an appropriate scaling of the rate functions together with the initial conditions (a large excess of particles) entails that $\{f^{\varepsilon}\}$ converges to a solution of the LS model Eq. (1.4). Here, we use the same strategy to construct solutions to LS and to derive appropriate flux conditions at $x=0$ when the reaction rates behave near 0 as a power-law, that is

$$a(x)\sim_{0^+}\overline{a}x^{r_a} \text{ and } b(x)\sim_{0^+}\overline{b}x^{r_b},$$

with $\overline{a}$ and $\overline{b}$ are positive numbers, and the exponents satisfying $0\leq r_a<1$ and $r_a\leq r_b$ which corresponds to entrant characteristic whenever $u(t)>\lim_{x\to 0^+}\frac{\overline{b}}{\overline{a}}x^{r_b-r_a}$.

Remark 1. *Another scaling approach considers the large time behavior of the Becker-Döring model, and relates the dynamics of large clusters to solutions of various version of Lifshitz-Slyozov equations. It is the so-called theory of Ostwald ripening, see* [23,27,30].

We emphasize that the novelty of our work is based on the rigorous derivation of a boundary condition at $x=0$ for the LS model Eq. (1.4), which is needed in the case of entrant characteristic. Thanks to new estimates on the BD model (Proposition 2), we identify the limit of quantities related to the (finite size) c_i^{ε}'s by a quasi steady state approximation. From this identification, we were able to found various possible boundary conditions depending on different scaling hypotheses on the first fragmentation rate, *i.e.* according to the value of η in (1.2), with respect to r_a and r_b. Namely, we found

three distinct cases for *slow* de-nucleation rate ($\eta > r_a$) in Theorem 1, *compensated* one ($\eta = r_a$) in Theorem 2 and *fast* one ($\eta < r_a$) in Theorem 3. We obtained these main results for measure-valued solution to the LS equation, in Section 3. But in Section 6, we improve these results to obtain density solutions when a and b are exact power laws. Let us give an example of the results obtained.

ILLUSTRATING RESULT. *Assume, for all $x \geq 0$, $a(x) = \overline{a}x^{r_a}$ and $b(x) = \overline{b}x^{r_b}$ with $r_a < r_b$ and $\eta = r_b$. We found the limit of $\{f^\varepsilon\}$ is a solution of Eq.* (1.4)*, with the boundary value given by, for all $t \geq 0$,*

$$\lim_{x \to 0^+} (a(x)u(t) - b(x))f(t,x) = \alpha u(t)^2,$$

where α is the limit of α^ε in (1.2)*. In other words we recover the behavior of f near $x = 0$ with the free particles' concentration through the limit*

$$\lim_{x \to 0^+} x^{r_a} f(t,x) = \frac{\alpha}{\overline{a}} u(t).$$

Organization of the paper. In the next Section 2 we introduce the main assumptions along with some properties of the BD model. Then, in Section 3 we state our main results on measure-valued solution to LS with boundary term. To do so we improved previous compactness arguments on the re-scaled density (1.6), so that the boundary term can be taken into account in Section 4. It is achieved thanks to a new estimate on the growth of the "small" sized clusters (point-wise estimates of the density approximation, see Proposition 2). The identification of the boundary term in Section 5 follows from a rigorous quasi-steady-state approximation of the small-sized clusters, in analogy with slow-fast systems, and allow proving the main theorems. Finally, we extend some results to a convergence in density, see Section 6. We conclude by a discussion and further directions in Section 7.

Notations. For any interval $I \subseteq \mathbb{R}$, we denote by $\mathcal{C}(I)$, respectively $\mathcal{C}_c(I)$ and $\mathcal{C}_b(I)$, the space of continuous function on I, respectively with compact support in I and bounded on I. We also denote by $\mathcal{C}_0(I)$ the completion of $\mathcal{C}_c(I)$ for the uniform norm. We denote by $\mathcal{M}_f(I)$ the cone of non-negative and finite regular Borel measures on I identifies, by the Riesz's representation theorem to the positive continuous linear form on $\mathcal{C}_0(I)$. We equip $\mathcal{M}_f(I)$ with the topology of the $weak-*$ convergence (sometimes called *vague*), *i.e.* for $\{\nu^\varepsilon\}$ and ν in $\mathcal{M}_f(I)$, ν^ε converges to ν in $\mathcal{M}_f(I)$ (in the $weak-*$ topology) if and only if for all $\varphi \in \mathcal{C}_0(I)$

$$\int_0^\infty \varphi(x)\nu^\varepsilon(dx) \to \int_0^\infty \varphi(x)\nu(dx).$$

Note for further remark that $\mathcal{M}_f(I)$ with the $weak-*$ topology is a completely metrizable space.

2. Preliminaries and Assumptions

In this section, we recall some known results on the BD system, together with assumptions for the main results of this paper. First of all, we refer the reader to Theorem 2.1 in [20] for existence and uniqueness of (non-negative) global solution to Eq. (1.1) satisfying the balance of mass Eq. (1.3) at fixed $\varepsilon > 0$. Well-posedness follows from growth conditions on the kinetic rates, namely we assume the following.

Assumption 1. *The rates α^ε, β^ε, $(a_i^\varepsilon)_{i\geq 2}$ and $(b_i^\varepsilon)_{i\geq 3}$ are positives and, for each $\varepsilon>0$, there exists a constant $K(\varepsilon)>0$ such that*

$$a_{i+1}^\varepsilon - a_i^\varepsilon \leq K(\varepsilon),\ i\geq 2,$$
$$b_i^\varepsilon - b_{i+1}^\varepsilon \leq K(\varepsilon),\ i\geq 3.$$

From now, for each $\varepsilon>0$, we assume u^ε and $(c_i^\varepsilon)_{i\geq 2}$ are given by the non-negative solution to (1.1), that belongs (each) to $\mathcal{C}([0,+\infty))$.

We construct aggregation and fragmentation rates as functions on $\mathbb{R}_+$ similarly to f^ε, namely, for each $\varepsilon>0$ we define, for all x in $\mathbb{R}_+$,

$$a^\varepsilon(x) := \sum_{i\geq 2} a_i^\varepsilon \mathbf{1}_{\Lambda_i^\varepsilon}(x),\ \text{and}\ b^\varepsilon(x) := \sum_{i\geq 3} b_i^\varepsilon \mathbf{1}_{\Lambda_i^\varepsilon}(x).$$

Now, we are able to derive a weak equation on the density approximation f^ε, for each $\varepsilon>0$, in which we will pass to the limit to recover weak solutions to Eq. (1.4). This next proposition follows from [20, Lemma 4.1].

Proposition 1. *Under Assumption 1, let $\{f^\varepsilon\}$ constructed by Eq. (1.6). For each $\varepsilon>0$, and all $\varphi\in W^{1,\infty}_{loc}(\mathbb{R}_+)$ such that $\partial_x\varphi\in L^\infty(\mathbb{R}_+)$, we have, for all $t\geq 0$,*

$$\begin{aligned}\int_0^{+\infty} & f^\varepsilon(t,x)\varphi(x)\,dx \\ &= \int_0^{+\infty} f^{in,\varepsilon}(x)\varphi(x)\,dx + \int_0^t [\alpha^\varepsilon u^\varepsilon(s)^2 - \beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(s)]\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon}\varphi(x)\,dx\right)ds \\ &\quad + \int_0^t\int_0^{+\infty} [a^\varepsilon(x)u^\varepsilon(s)f^\varepsilon(s,x)\Delta_\varepsilon\varphi(x) - b^\varepsilon(x)f^\varepsilon(s,x)\Delta_{-\varepsilon}\varphi(x)]\,dx\,ds, \qquad (2.1)\end{aligned}$$

where $\Delta_h\varphi(x) = (\varphi(x+h)-\varphi(x))/h$, for $h\in\mathbb{R}$, and

$$u^\varepsilon(t) + \int_0^\infty x f^\varepsilon(t,x)\,dx = m^\varepsilon. \qquad (2.2)$$

In the next assumption we assume standard hypotheses on the convergence of the rate functions and their sub-linear control, see also [9, 20].

Assumption 2. (Convergence of the rates). *Let α and β be two positive numbers, and let a and b be two non-negative continuous functions on $[0,+\infty)$ that are positive on $x\in(0,+\infty)$. As $\varepsilon\to 0$, we suppose that*

$\{\alpha^\varepsilon\}$ *converges towards* α. (H1)

$\{\beta^\varepsilon\}$ *converges towards* β. (H2)

$\{a^\varepsilon(.)\}$ *converges uniformly on any compact set of* $[0,+\infty)$ *towards* $a(.)$ *and* $\exists K_a>0$ *s.t.* $a^\varepsilon(x)\leq K_a(1+x)$, $\forall x\in\mathbb{R}_+$ *and* $\forall\varepsilon>0$. (H3)

$\{b^\varepsilon(.)\}$ *converges uniformly on any compact set of* $[0,+\infty)$ *towards* $b(.)$ *and* $\exists K_b>0$ *s.t.* $b^\varepsilon(x)\leq K_b(1+x)$, $\forall x\in\mathbb{R}_+$ *and* $\forall\varepsilon>0$. (H4)

We recall a discussion on the scaling of the coefficients is differed to Section 7.

The next assumption details the behavior of the rate functions around 0. This is the essential assumption which allows us to identify the limit of $\varepsilon^\eta c_2^\varepsilon$ in the second integral in the right hand side of Eq. (2.1).

Assumption 3. (Behavior of the rate functions near 0). *We suppose there exist* $r_a \in [0,1)$, $r_b \geq r_a$, $\overline{a}>0$, $\overline{b}>0$ *such that*

$$\begin{array}{l|l} a(x) \sim_{0^+} \overline{a} x^{r_a}, & b(x) \sim_{0^+} \overline{b} x^{r_b}, \\ a^\varepsilon(\varepsilon i) = a(\varepsilon i) + o((\varepsilon i)^{r_a}), & b^\varepsilon(\varepsilon i) = b(\varepsilon i) + o((\varepsilon i)^{r_b}), \end{array} \tag{H5}$$

where o is the Landau notation, i.e. $o(x)/x \to 0$ as $x \to 0$. We define the quantity

$$\rho := \lim_{x\to 0^+} \frac{b(x)}{a(x)} = \lim_{x\to 0^+} \frac{\overline{b}}{\overline{a}} x^{r_b - r_a} \in [0,+\infty). \tag{2.3}$$

In the case $r_a = r_b$ ($\rho > 0$), we assume moreover that

$$b(x) \geq \rho a(x). \tag{2.4}$$

Remark 2. *Note, if $0 \leq r_b < r_a$ or $r_a \geq 1$, the kinetic rates a and b are related to outgoing characteristics for which the theory already exists, see [9, 20]. The quantity ρ defined above determines whether the characteristics at $x=0$ are ongoing or outgoing, according to whether $u(t)$ is greater or less than ρ. The assumption given by the relation (2.4) is a technical condition needed to ensure that if u starts above the critical threshold ρ, it stays above for all times (see lemma 7). Removing this hypothesis would provide only local in time convergence results in Theorems 1, 2, 3 when $r_a = r_b$.*

Finally, we assume some control on the initial conditions. For this, we introduce a set of functions which shall play a key role. We denote by $\mathcal{U}$ the set of non-negative convex functions Φ belonging to $\mathcal{C}^1([0,+\infty))$ and piecewise $\mathcal{C}^2([0,+\infty))$ such that $\Phi(0)=0$, Φ' is concave, $\Phi'(0) \geq 0$, and

$$\lim_{x\to+\infty} \frac{\Phi(x)}{x} = +\infty.$$

Note that Φ is increasing. These functions have remarkable properties when conjugate to the structure of the Becker-Döring system and provide important estimates, see for instance [19].

Assumption 4. (Initial conditions). *We assume there exists $u^{\text{in}} > \rho$ and a non-negative measure $\mu^{\text{in}} \in \mathcal{M}_f([0,+\infty))$ such that $u^{\text{in},\varepsilon}$ converges to u^{in} in $\mathbb{R}_+$ and $\{f^{\text{in},\varepsilon}\}$ converges to μ^{in}, in the weak $-*$ topology of $\mathcal{M}_f([0,+\infty))$. Moreover, we assume there exists $\Phi \in \mathcal{U}$ such that*

$$\sup_{\varepsilon>0} \int_0^\infty \Phi(x) f^{\text{in},\varepsilon}(x) dx < +\infty. \tag{H6}$$

In particular, we can define

$$m := u^{\text{in}} + \int_0^\infty x \mu^{\text{in}}(dx).$$

Moreover, we suppose that for all $z \in (0,1)$,

$$\sup_{\varepsilon>0} \sum_{i\geq 2} \varepsilon^{r_a} c_i^{\mathrm{in},\varepsilon} e^{-iz} < +\infty. \tag{H7}$$

REMARK 3. *m is well-defined since weak$-*$ convergence plus the extra-moment in* (H6) *give the limit*

$$\int_0^\infty x f^{\mathrm{in},\varepsilon}(dx) \to \int_0^\infty x \mu^{\mathrm{in}}(dx).$$

See for instance [9, *Proof of Theorem 2.3*].

REMARK 4. *In fact, we could obtain freely this Φ assuming a stronger weak convergence (against $(1+x)\varphi(x)$ for φ bounded and continuous). See for instance* [7] *for the construction of such a Φ.*

REMARK 5. *We highlight that condition* (H7) *is not restrictive. For example, consider $f^{\mathrm{in}}(x) = x^{-r}$ on* $(0,1)$ *and* 0 *elsewhere, with $r \leq r_a$. Then, consider $c_i^{\mathrm{in},\varepsilon} = (i\varepsilon)^{-r}$ for $i \leq 1/\varepsilon$, and* 0 *elsewhere. We have that $\{f^{\mathrm{in},\varepsilon}\}$ trivially converges to f^{in} in the sense of* (H6) *and it satisfies* (H7). *Note that we do not necessarily require the initial condition is composed of " very large" clusters (of size $i \gg 1/\varepsilon$).*

3. Main results

For the remainder of the paper, we always assume that $\{f^\varepsilon\}$ is constructed by Eq. (1.6), that $\{u^\varepsilon\}$ is given by the balance (2.2), and Assumption 1 to Assumption 4 hold true. The next definition extends the notion of a solution to the LS model Eq. (1.4), with a general boundary condition, or *nucleation rate*.

DEFINITION 1. N-solution. *Let $T > 0$, a function $N \in L^\infty_{loc}(\mathbb{R}_+)$ called nucleation rate, $u^{\mathrm{in}} > \rho$, a measure $\mu^{\mathrm{in}} \in \mathcal{M}_f([0,+\infty))$, and a measure-valued function $\mu \in L^\infty([0,T]; w-*-\mathcal{M}_f([0,+\infty))$. We say that μ is a N-solution to the LS equation (in measure) on $[0,T]$ with mass m, when:*

i) There exists a non-negative $u \in \mathcal{C}([0,T])$, such that $u(0) = u^{\mathrm{in}}$, and for all $t \in [0,T]$,

$$u(t) + \int_0^\infty x \mu_t(dx) = m. \tag{3.1}$$

ii) For all $\varphi \in \mathcal{C}^1_c([0,T) \times [0,+\infty))$

$$\int_0^T \int_0^\infty \left[\partial_t \varphi(t,x) + (a(x)u(t) - b(x))\partial_x \varphi(t,x)\right] \mu_t(dx)\, dt + \int_0^\infty \varphi(0,x)\mu^{\mathrm{in}}(dx) + \int_0^T \varphi(s,0) N(u(s))\, ds = 0, \tag{3.2}$$

REMARK 6. *The space $L^\infty([0,T]; w-*-\mathcal{M}_f([0,+\infty))$ consists of (equivalent classes of) measurable functions from $(0,T)$ to $\mathcal{M}_f([0,+\infty))$ with respect to the weak$-*$ topology that are essentially bounded,*

$$\operatorname*{ess\,sup}_{t\in(0,T)} \mu_t([0,\infty)) < +\infty.$$

The essential supremum defines a norm on this space.

We now state the main results. The first theorem, when $\eta > r_a$, corresponds to the case where the first fragmentation rate is too slow and does not contribute to the boundary value. Thus, the nucleation rate is proportional to the number of encounter of free particles, namely $u(t)^2$ at time t.

THEOREM 1. The *slow* de-nucleation case. *Assume $\eta > r_a$. For any $T > 0$ and any sequence $\{\varepsilon_n\}$ converging to 0, it exists a sub-sequence $\{\varepsilon_{n'}\}$ of $\{\varepsilon_n\}$ and μ a N-solution to LS on $[0,T]$ with mass m, such that*

$$f^{\varepsilon_{n'}} \xrightarrow[n' \to +\infty]{} \mu$$

in $\mathcal{C}([0,T]; w-*-\mathcal{M}_f([0,+\infty))$, where, for all $u \geq 0$

$$N(u) = \alpha u^2.$$

REMARK 7. *The space $\mathcal{C}([0,T]; w-*-\mathcal{M}_f([0,+\infty)))$ has to be understood as the measure-valued functions that are continuous in time for the weak$-*$ topology on $\mathcal{M}_f([0,+\infty))$, i.e. for $\{\nu_t\} \in \mathcal{C}([0,T]; w-*-\mathcal{M}_f([0,+\infty)))$, we have, for all $t \in [0,T]$ and $\varphi \in \mathcal{C}_0([0,+\infty))$,*

$$t \mapsto \int_0^\infty \varphi(x)\nu_t(dx)$$

is continuous. This space is equipped with the uniform topology (note that $w-*-\mathcal{M}_f([0,+\infty))$ is metrizable).

The second theorem holds in the limit case when $\eta = r_a$, *i.e.* the first fragmentation rate has the same order of magnitude than the aggregation rate ($i \geq 2$). Compared to the first case, the nucleation rate is balanced by a function varying between 0 and 1.

THEOREM 2. The *compensated* de-nucleation case. *Assume $\eta = r_a$. For any $T > 0$ and any sequence $\{\varepsilon_n\}$ converging to 0, it exists a sub-sequence $\{\varepsilon_{n'}\}$ of $\{\varepsilon_n\}$ and μ a N-solution to LS on $[0,T]$ with mass m, such that*

$$f^{\varepsilon_{n'}} \xrightarrow[n' \to +\infty]{} \mu$$

in $\mathcal{C}([0,T]; w-*-\mathcal{M}_f([0,+\infty))$, where, for all $u \geq 0$

$$N(u) = \begin{cases} \alpha u^2 \dfrac{u}{u + \beta/(\bar{a}2^\eta)}, & \text{if } \eta = r_a < r_b, \\ \alpha u^2 \dfrac{\bar{a}u - \bar{b}}{\bar{a}u - \bar{b} + \beta/2^\eta}, & \text{if } \eta = r_a = r_b, \end{cases}$$

REMARK 8. *In the pure aggregation case, with $\beta^\varepsilon = b_i^\varepsilon = 0$, then $b = 0$ and $\beta = \bar{b} = 0$. Our results in Theorem 1 and Theorem 2 are consistent and remain true.*

REMARK 9. *In both Theorems 1 and 2, the continuity in time of the limit allows to recover the moment solutions, that is for all $\varphi \in \mathcal{C}([0,+\infty))$ and $t \in [0,T]$*

$$\int_0^\infty \varphi(x)\mu_t(dx) = \int_0^\infty \varphi(x)\mu^{\text{in}}(dx) + \int_0^t \int_0^\infty (a(x)u(s) - b(x))\varphi'(x)\mu_s(dx)\,ds + \int_0^t \varphi(0)N(u(s))\,ds = 0,$$

Finally, the last theorem considers the case of a fast de-nucleation rate so that the flux at the boundary vanishes, and the solution can reveal fast oscillations or discontinuities in time near $x=0$.

THEOREM 3. The *fast* de-nucleation rate. *Assume $\eta<r_a$. For any $T>0$ and any sequence $\{\varepsilon_n\}$ converging to 0, it exists a sub-sequence $\{\varepsilon_{n'}\}$ of $\{\varepsilon_n\}$ and μ a N-solution to LS on $[0;T]$ with mass m, such that the restriction of μ to $(0,+\infty)$ is $\mathcal{C}([0,T];w-*-\mathcal{M}_f((0,+\infty))$ and*

$$f^{\varepsilon_{n'}} \xrightarrow[n'\to+\infty]{} \mu$$

in weak$-*-L^\infty([0,T];w-*-\mathcal{M}_f([0,+\infty))$, where, for all $u\geq 0$

$$N(u)=0.$$

REMARK 10. *Since $\mathcal{C}_0([0,+\infty))$ is a separable Banach space (and since the Lebesgue measure on $[0,T]$ is σ-finite) we can identify $L^\infty([0,T];w-*-\mathcal{M}_f([0,+\infty))$ with the positive continuous linear forms on $L^1([0,T];\mathcal{C}_0([0,+\infty))$ whose (operator) norm is given by the essential supremum defined in Remark 6, see* [13, *Section 8.18*] *and* [4, *Chap. 6, §2, n. 6, Proposition 10*]. *Thus, this space can be equipped with the classical weak$-*$ topology for which we derived convergence in the theorems.*

REMARK 11. *In the last case, for Theorem 3, we were not able to prove equicontinuity of the density approximation in $w-*-\mathcal{M}_f([0,+\infty))$. But, equicontinuity remains true when the measure is restricted to $(0,+\infty)$, open in $x=0$. This suggests the limit solution is more regular than only L^∞ in time. Thus, fast oscillations or discontinuities in time can only occur at $x=0$.*

4. Compactness estimates

In this section we provide the main estimates to obtain sufficient compactness arguments to pass to the limit in (2.1)-(2.2). Remark for further estimations, under (H1) and (H2), there exists a positive $K_{\alpha,\beta}$ such that, for all $\varepsilon>0$,

$$\alpha^\varepsilon,\beta^\varepsilon,\alpha,\beta\in(0,K_{\alpha,\beta}], \tag{4.1}$$

and (H3)-(H4) imply that the limit functions also satisfy

$$a(x)\leq K_a(1+x) \text{ and } b(x)\leq K_b(1+x),\ \forall x\in[0,+\infty). \tag{4.2}$$

We fix these constants for the remainder.

4.1. Uniform bound for the density approximation

The first lemma gives basic estimates. In particular, it constructs the compact set of $\mathcal{M}_f([0,+\infty))$ in which the sequence of solutions remains.

LEMMA 1. *For all $T>0$,*

$$\sup_{\varepsilon>0}\sup_{t\in[0,T]}\int_0^{+\infty}(1+x+\Phi(x))f^\varepsilon(t,x)\,dx<+\infty, \tag{4.3}$$

$$\sup_{\varepsilon>0}\sup_{t\in[0,T]}u^\varepsilon(t)<+\infty, \tag{4.4}$$

$$\sup_{\varepsilon>0}\int_0^T\varepsilon^\eta c_2^\varepsilon(t)\,dt<+\infty. \tag{4.5}$$

Remark 12. *Similar estimates can be found in* [20] *for a different scaling. For sake of completeness we recall the proof below. Note that estimate* (4.5)*, although trivial, seems to have not been reported elsewhere, and will be important for the next.*

Proof. By Assumption 4, the convergence of $\{f^{\mathrm{in},\varepsilon}\}$ implies that the sequence lies in a *weak*$-*$ compact set of $\mathcal{M}_f([0+\infty))$, and with (H6) we have

$$\sup_{\varepsilon>0}\int_{\mathbb{R}_+} f^{in,\varepsilon}(x)(1+x+\Phi(x))dx<+\infty. \tag{4.6}$$

Let us start now with the estimate (4.4). By the mass conservation relationship (2.2), $u^\varepsilon(t)\le m^\varepsilon$, for any $t\ge 0$, and thanks to Assumption 4, (m^ε) converges as $\varepsilon\to 0$, thus it is bounded by a constant $K_m>0$. Then estimate (4.4) directly follows. Similarly, we obtain

$$\sup_{\varepsilon>0}\sup_{t\in[0,T]}\int_0^{+\infty} xf^\varepsilon(t,x)\,dx<+\infty.$$

Then, taking $\varphi=\mathbf{1}$ in Eq. (2.1), it immediately yields by re-arranging the non-positive terms

$$0\le\int_0^{+\infty} f^\varepsilon(t,x)\,dx+\int_0^t \beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(s)\,ds\le\int_0^{+\infty} f^{in,\varepsilon}(x)\,dx+\int_0^t \alpha^\varepsilon u^\varepsilon(s)^2\,ds.$$

Using bounds (4.1), (4.4) and (4.6), we obtain the inequality (4.5) together with the first part of estimate (4.3). Finally, we put $\varphi=\Phi$ in Eq. (2.1). Remark that the derivative Φ' is not uniformly bounded, thus we cannot use Proposition 1 straightforwardly. However, with a classical regularizing argument, one can show that the next computations hold true *a posteriori*, see for instance [20, proof of Lemma 4.2]. We remark that

$$0\le\Delta_\varepsilon\Phi(x)\le\Phi'(x+\varepsilon),\quad -\Delta_{-\varepsilon}\Phi(x)\le-\Phi'(x)\le 0.$$

Moreover, $\Phi'(x+\varepsilon)\le\Phi'(x)+\varepsilon\Phi''(0)$. Thus, dropping the non-positive terms, using (H3) and again that $u^\varepsilon(t)\le K_m$,

$$\begin{aligned}\int_0^{+\infty} f^\varepsilon(t,x)\Phi(x)\,dx\le&\int_0^{+\infty} f^{in,\varepsilon}(x)\Phi(x)\,dx+\int_0^t \alpha^\varepsilon u^\varepsilon(s)^2\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon}\Phi(x)dx\right)ds\\&+K_mK_a\int_0^t\int_0^{+\infty}(1+x)f^\varepsilon(s,x)(\Phi'(x)+\varepsilon\Phi''_{1,r}(0))\,dx\,ds,\end{aligned}\tag{4.7}$$

Let $\delta>0$. Note that $x\Phi'(x)\le 2\Phi(x)$ by [18, Lemma A.1], thus we get

$$\begin{aligned}\int_0^{+\infty}(1+x)f^\varepsilon(s,x)\Phi'(x)\,dx&\le\int_0^\delta f^\varepsilon(s,x)\Phi'(x)\,dx+\left(\frac{1}{\delta}+1\right)\int_0^{+\infty} xf^\varepsilon(s,x)\Phi'(x)\,dx\\&\le\big(\sup_{(0,\delta)}\Phi'\big)\int_0^\infty f^\varepsilon(s,x)\,dx+2\left(\frac{1}{\delta}+1\right)\int_0^{+\infty} f^\varepsilon(s,x)\Phi(x)\,dx.\end{aligned}$$

We introduce this last estimate into Eq. (4.7) and we conclude using previous bounds and Grönwall's lemma. □

4.2. Pointwise estimations on the density

We turn now to the main estimate of this paper. Indeed, to obtain equicontinuity for the density $\{f^\varepsilon\}$ (in a measure space), and then identify the boundary condition, we need to control the behavior of the small-sized clusters, particularly because of the term $\varepsilon^\eta c_2^\varepsilon$ in the weak equation (2.1). Remark that we already have a weak bound (in time) given by Eq. (4.5). In the next Proposition 2 we improve this estimate by a control on exponential moments which depends on ρ (defined in Eq. (2.3)). Moments are classical tools and play a key role in the well-posedness of BD theory. More recently, exponential moments were also used [6, 16] to study long time behavior of BD solutions. Here, let us define the discrete Laplace transform

$$F^\varepsilon(t,z)=\sum_{j\geq 2}\varepsilon^{r_a}c_j^\varepsilon(t)e^{-jz},\quad z\in(0,1). \tag{4.8}$$

From the re-scaled system (1.1), the sequence $(d_i^\varepsilon)_{i\geq 2}$ defined by $d_i^\varepsilon:=\varepsilon^{r_a}c_i^\varepsilon$, for $i\geq 2$, satisfies, for each $\varepsilon>0$, the following equations

$$\varepsilon^{1-r_a}\frac{d}{dt}d_i^\varepsilon(t)=H_{i-1}^\varepsilon-H_i^\varepsilon,\quad i\geq 2, \tag{4.9}$$

where the fluxes are

$$H_1^\varepsilon=\alpha^\varepsilon u^\varepsilon(t)^2-\beta^\varepsilon\varepsilon^{\eta-r_a}d_2^\varepsilon(t),\ \text{and}\ H_i^\varepsilon=\overline{a}_i^\varepsilon u^\varepsilon(t)d_i^\varepsilon(t)-\varepsilon^{r_b-r_a}\overline{b}_{i+1}^\varepsilon d_{i+1}^\varepsilon(t),\quad i\geq 2,$$

with, for all $i\geq 2$,

$$\overline{a}_i^\varepsilon=\frac{a_i^\varepsilon}{\varepsilon^{r_a}},\quad\text{and}\quad \overline{b}_{i+1}^\varepsilon=\frac{b_{i+1}^\varepsilon}{\varepsilon^{r_b}}.$$

Note that, under hypotheses (H3), (H4) and (H5), the kinetic coefficients α^ε, β^ε and $\overline{a}_i^\varepsilon$, $\overline{b}_i^\varepsilon$, $i\geq 2$, are convergent sequences toward a positive value (resp. α, β, $\overline{a}i^{r_a}$, $\overline{b}i^{r_b}$).

Proposition 2. *Let $T>0$ and $\{\varepsilon_n\}$ a sequence converging to 0 such that $\{u^{\varepsilon_n}\}$ converges toward u uniformly on $[0,T]$, with $\inf_{t\in[0,T]}u(t)>\rho$. There exists $z_0>0$ such that for all $z\in(0,z_0)$*

$$\sup_{n\geq 0}\sup_{t\in[0,T]}F^{\varepsilon_n}(t,z)<\infty. \tag{4.10}$$

In particular, for all $r\geq r_a$ and $i\geq 2$, we have

$$\sup_{n\geq 0}\sup_{t\in[0,T]}\varepsilon^r c_i^{\varepsilon_n}(t)<+\infty. \tag{4.11}$$

Remark 13. *It is immediate from estimate (4.11) that we can obtain compactness in $w-*-L^\infty(0,T)$ for any finite size cluster $\varepsilon^r c_i^\varepsilon$, which will be used to prove both theorems 1 and 2.*

Remark 14. *We cannot prove that the pseudo-moment F^ε is propagated along limit solution for which $u(t)\leq\rho$ on some time interval. This is important in the case $r_a=r_b$ since $\rho>0$ and u can eventually cross this threshold, which is up to our knowledge an open problem. For that reason, we imposed the extra assumption (2.4) on a,b, needed in the proof of lemma 7.*

Proof. Let $z>0$ and $\varepsilon>0$. First, note the discrete Laplace transform defined in Eq. (4.8) is finite for each $\varepsilon>0$ and for all t in $[0,T]$, since

$$F^\varepsilon(t,x)\le \varepsilon^{r_a-1}\int_0^\infty x f^\varepsilon(t,x)\,dx.$$

Let us derive F^ε with respect to time (derivation under the sum is justified by similar bound). For all $t\in[0,T]$, we get

$$\varepsilon^{1-r_a}\partial_t F^\varepsilon(t,z)=\sum_{j\ge 2}e^{-jz}\left[H^\varepsilon_{j-1}-H^\varepsilon_j\right]=e^{-2z}H^\varepsilon_1-(1-e^{-z})\sum_{j\ge 2}e^{-jz}H^\varepsilon_j.$$

Thus, after developing the fluxes we get

$$\begin{aligned}\varepsilon^{1-r_a}\partial_t F^\varepsilon(t,z)=e^{-2z}H^\varepsilon_1-(1-e^{-z})\sum_{j\ge 2}e^{-jz}\overline{a}^\varepsilon_j u^\varepsilon(t)d^\varepsilon_j(t)\\ +(1-e^{-z})\sum_{j\ge 2}e^{-jz}\varepsilon^{r_b-r_a}\overline{b}^\varepsilon_{j+1}d^\varepsilon_{j+1}(t).\end{aligned}$$

Then, re-indexing the second sum on the right hand side, we obtain

$$\begin{aligned}\varepsilon^{1-r_a}\partial_t F^\varepsilon(t,z)=e^{-2z}H^\varepsilon_1-(1-e^{-z})e^{-2z}\overline{a}^\varepsilon_2 u^\varepsilon(t)d^\varepsilon_2(t)\\ -(1-e^{-z})\sum_{j\ge 3}e^{-jz}\overline{a}^\varepsilon_j\left[u^\varepsilon(t)-\frac{b^\varepsilon_j}{a^\varepsilon_j}e^z\right]d^\varepsilon_j(t).\end{aligned}\tag{4.12}$$

Since $\inf_{t\in[0,T]}u(t)>\rho$, we can find a constant c such that $\inf_{t\in[0,T]}u(t)\ge c>\rho$. Then, by uniform convergence of $\{u^{\varepsilon_n}\}$, there exists $\tilde\varepsilon>0$ small enough, such that for all n with $\varepsilon_n\le\tilde\varepsilon$, $\inf_{t\in[0,T]}u^{\varepsilon_n}(t)\ge c>\rho$. Also, we can choose $\delta>0$ and $z_0>0$, both small enough, such that for all $t\in[0,T]$ we have $c>\rho e^{z_0}+2\delta$. Then, there exists $N>0$ such that, for all $z\in(0,z_0)$

$$\inf_{n\ge N}\inf_{t\in[0,T]}u^{\varepsilon_n}(t)>\rho e^z+2\delta.$$

Then, by hypothesis (H5), for all $3\le j\le 1/\sqrt{\varepsilon}$,

$$\frac{b^\varepsilon_j}{a^\varepsilon_j}=\frac{\overline{b}}{\overline{a}}\frac{(\varepsilon j)^{r_b}+o((\varepsilon j)^{r_b})}{(\varepsilon j)^{r_a}+o((\varepsilon j)^{r_a})}=\frac{\overline{b}}{\overline{a}}(\varepsilon j)^{r_b-r_a}(1+o(1)),$$

so that, we have, for N large enough,

$$\sup_{n\ge N}\sup_{j\in[3,\dots,\lfloor 1/\sqrt{\varepsilon_n}\rfloor-1]}\left|\rho-\frac{b^{\varepsilon_n}_j}{a^{\varepsilon_n}_j}\right|<\delta e^{-z}.$$

The latter gives a uniform control in j for the relatively "small" sizes $j\le 1/\sqrt{\varepsilon}$. We separate the sum in Eq. (4.12) in two parts, the small-size clusters for $j\in(3,\dots,\lfloor 1/\sqrt{\varepsilon_n}\rfloor-1)$ in one side, for which (for $n\ge N$)

$$u^{\varepsilon_n}(t)-\frac{b^{\varepsilon_n}_j}{a^{\varepsilon_n}_j}e^z=u^{\varepsilon_n}(t)-\rho e^z+e^z\left(\rho-\frac{b^{\varepsilon_n}_j}{a^{\varepsilon_n}_j}\right)\ge 2\delta-\delta=\delta,$$

and the large-size clusters in another side. Hence, for all $t\in[0,T]$,

$$\sum_{j\geq 3} e^{-jz}\overline{a}_j^{\varepsilon_n}\left[u^{\varepsilon_n}(t)-\frac{b_j^{\varepsilon_n}}{a_j^{\varepsilon_n}}e^z\right]d_j^{\varepsilon_n}(t)$$
$$\geq \delta \sum_{j=3}^{\lfloor 1/\sqrt{\varepsilon_n}\rfloor -1} e^{-jz}\overline{a}_j^{\varepsilon_n} d_j^{\varepsilon_n}(t) + \sum_{j\geq \lfloor 1/\sqrt{\varepsilon_n}\rfloor} e^{-jz}\overline{a}_j^{\varepsilon_n}\left[u^{\varepsilon_n}(t)-\frac{b_j^{\varepsilon_n}}{a_j^{\varepsilon_n}}e^z\right]d_j^{\varepsilon_n}(t). \quad (4.13)$$

Using hypothesis (H5), there exists x_0 such that for all $x\in(0,x_0)$, $a(x)/x^{r_a}>3\overline{a}/4$. Thus, there exists $\tilde{N}$ such that for all $n\geq\tilde{N}$ and for all $2\leq i\leq 1/\sqrt{\varepsilon_n}$ we have $\varepsilon_n i\leq\sqrt{\varepsilon_n}<x_0$ and $a(\varepsilon i)/(\varepsilon_n i)^{r_a}\geq 3\overline{a}/4$. Still, with hypothesis (H5), we can choose $\tilde{N}$ such that for all $n>\tilde{N}$, and for all $2\leq i\leq 1/\sqrt{\varepsilon_n}$, we have $a^{\varepsilon}(\varepsilon i)/(\varepsilon_n i)^{r_a}\geq\overline{a}/2$. Hence, from the rank $\tilde{N}$, there exists a constant $\widetilde{K}_a>0$ such that for all $n\geq\tilde{N}$ and for all $2\leq j\leq 1/\sqrt{\varepsilon_n}$, we have

$$\overline{a}_j^{\varepsilon_n}=\frac{a_j^{\varepsilon_n}}{\varepsilon_n^{r_a}}\geq\widetilde{K}_a:=\frac{1}{2}\overline{a}2^{r_a}.$$

Accordingly, the rest of the proof has to be understood for n large enough. Using the equation on H_1^{ε} and plugging inequality (4.13) into Eq. (4.12) we obtain

$$\varepsilon_n^{1-r_a}\partial_t F^{\varepsilon_n}(t,z)\leq e^{-2z}[\alpha^{\varepsilon_n}u^{\varepsilon_n}(t)^2-\varepsilon_n^{\eta-r_a}\beta^{\varepsilon_n}d_2^{\varepsilon_n}(t)]$$
$$-(1-e^{-z})e^{-2z}[\overline{a}_2^{\varepsilon_n}u^{\varepsilon_n}(t)-\delta\tilde{K}_a]d_2^{\varepsilon_n}(t)-(1-e^{-z})\delta\widetilde{K}_a\sum_{j=2}^{\lfloor 1/\sqrt{\varepsilon_n}\rfloor -1}e^{-jz}d_j^{\varepsilon_n}(t)$$
$$-(1-e^{-z})\sum_{j\geq\lfloor 1/\sqrt{\varepsilon_n}\rfloor}e^{-jz}\overline{a}_j^{\varepsilon_n}\left[u^{\varepsilon_n}(t)-\frac{b_j^{\varepsilon_n}}{a_j^{\varepsilon_n}}e^z\right]d_j^{\varepsilon_n}(t).$$

We remark that $\overline{a}_2^{\varepsilon_n}u^{\varepsilon_n}(t)-\delta\widetilde{K}_a\geq\tilde{K}_a(\rho e^z+2\delta-\delta)\geq\widetilde{K}_a\rho\geq 0$. Using the moment estimates (4.4) and hypothesis (H3), we have $\sup_{t\in[0,T]}\alpha^{\varepsilon}u^{\varepsilon}(t)^2\leq K_0$ uniformly in $\varepsilon>0$. Thus, dropping also some negative terms, we have

$${\varepsilon_n}^{1-r_a}\partial_t F^{\varepsilon_n}(t,z)\leq K_0e^{-2z}-(1-e^{-z})\delta\widetilde{K}_a\sum_{j=2}^{\lfloor 1/\sqrt{\varepsilon_n}\rfloor -1}e^{-jz}d_j^{\varepsilon_n}(t)$$
$$+(1-e^{-z})\sum_{j\geq\lfloor 1/\sqrt{\varepsilon_n}\rfloor}e^{-jz}\frac{b_j^{\varepsilon_n}}{{\varepsilon_n}^{r_a}}d_j^{\varepsilon_n}(t).$$

Now using that

$$\sum_{j=2}^{\lfloor 1/\sqrt{\varepsilon_n}\rfloor -1}e^{-jz}d_j^{\varepsilon_n}(t)=F^{\varepsilon_n}(t,z)-\sum_{j\geq\lfloor 1/\sqrt{\varepsilon_n}\rfloor}e^{-jz}d_j^{\varepsilon_n}(t),$$

we obtain

$${\varepsilon_n}^{1-r_a}\partial_t F^{\varepsilon_n}(t,z)\leq K_0e^{-2z}-(1-e^{-z})\delta\widetilde{K}_aF^{\varepsilon_n}(t,z)$$
$$+(1-e^{-z})\delta\sum_{j\geq\lfloor 1/\sqrt{\varepsilon_n}\rfloor}e^{-jz}\widetilde{K}_ad_j^{\varepsilon_n}(t)+(1-e^{-z})e^z\sum_{j\geq\lfloor 1/\sqrt{\varepsilon_n}\rfloor}e^{-jz}\frac{b_j^{\varepsilon_n}}{{\varepsilon_n}^{r_a}}d_j^{\varepsilon_n}(t).$$

At this point, we recall that by definition we have, for all $j \geq 2$, $d_j^\varepsilon/\varepsilon^{r_a} = c_j^\varepsilon$, and $\widetilde{K}_a < a_j^\varepsilon/\varepsilon^{r_a}$, so that, with $K = \max(\delta, e^z)$,

$$\varepsilon_n{}^{1-r_a}\partial_t F^{\varepsilon_n}(t,z) \leq K_0 e^{-2z} - (1-e^{-z})\delta\widetilde{K}_a F^{\varepsilon_n}(t,z) + (1-e^{-z})K \sum_{j \geq \lfloor 1/\sqrt{\varepsilon_n}\rfloor} e^{-jz}(a_j^{\varepsilon_n} + b_j^{\varepsilon_n})c_j^{\varepsilon_n}(t).$$

Finally, by hypotheses (H3)-(H4), we have, for all $j \geq \lfloor 1/\sqrt{\varepsilon}\rfloor$ (and ε small enough)

$$e^{-jz}(a_j^\varepsilon + b_j^\varepsilon) \leq (K_a + K_b)(1+\varepsilon j)e^{-jz} \leq (K_a + K_b)\varepsilon.$$

Thus,

$$\varepsilon_n{}^{1-r_a}\partial_t F^{\varepsilon_n}(t,z) \leq K_0 e^{-2z} - (1-e^{-z})\delta\widetilde{K}_a F^{\varepsilon_n}(t,z) + (1-e^{-z})K(K_a + K_b)\int_0^{+\infty} f^{\varepsilon_n}(t,x)dx.$$

By the moment estimates (4.3), there exists $\tilde{K}$ independent from ε_n such that

$$\varepsilon_n{}^{1-r_a}\partial_t F^{\varepsilon_n}(t,z) \leq -(1-e^{-z})\delta\widetilde{K}_a F^{\varepsilon_n}(t,z) + \tilde{K}. \tag{4.14}$$

We can conclude that

$$F^{\varepsilon_n}(t,z) \leq F^{\varepsilon_n}(0,z) + \frac{\widetilde{K}}{\delta\widetilde{K}_a(1-e^{-z})},$$

and the result (4.10) follows thanks to the initial bound on $F^\varepsilon(0,z)$ given by hypothesis (H7). Note that (4.11) directly follows from the previous bound (4.10) and the definition of the discrete Laplace transform (4.8). □

Remark 15. *The estimate* (4.14) *on* F^ε *can be easily generalized for any exponent* r *instead of* r_a. *Writing* $G^\varepsilon(t,z) = \sum_{j\geq 2}\varepsilon^r c_j^\varepsilon(t)e^{-jz}$, *and following the same steps, we find*

$$\varepsilon^{1-r_a}\partial_t G^\varepsilon(t,z) \leq -(1-e^{-z})\delta\widetilde{K}_a G^\varepsilon(t,z) + \varepsilon^{r-r_a}\tilde{K}.$$

Thus, this inequality provides valuable information if $r \geq r_a$.

4.3. Equicontinuity lemmas

We now turn to the equicontinuity of the density approximation, as a measure valued time-dependent function. The new result here is to provide equicontinuity in a measure space on $[0,\infty)$ (see Lemma 4) . The first lemma is independent on η and similar to [9, 20].

Lemma 2. *Let* $T > 0$. *The family* $\{u^\varepsilon\}$ *is equicontinuous on* $[0,T]$.

Proof. Let us fix $T > 0$. From the mass conservation (2.2), we can deduce that the equicontinuity of $\{u^\varepsilon\}$ directly follows from the one of the sequence $\{\int_0^{+\infty} x f^\varepsilon(\cdot,x)dx\}$. Thus, we focus on the latter. We have, from Eq. (2.1) with $\varphi(x) = x$, for all $t \in [0, T-h]$ and $s \in [0,h]$ with $0 < h < T$,

$$\left|\int_0^{+\infty}[f^\varepsilon(t+s,x) - f^\varepsilon(t,x)]x\,dx\right| \leq \left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon} x\,dx\right)\int_t^{t+s}(\alpha^\varepsilon u^\varepsilon(\sigma)^2 + \beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(\sigma))\,d\sigma + \int_t^{t+s}\int_0^{+\infty}|a^\varepsilon(x)u^\varepsilon(\sigma)f^\varepsilon(\sigma,x) - b^\varepsilon(x)f^\varepsilon(\sigma,x)|\,dx\,d\sigma. \tag{4.15}$$

The first term in the r.h.s of (4.15) can be bounded, thanks to the bound (4.1), by

$$\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon} x\,dx\right)\int_t^{t+s}(\alpha^\varepsilon u^\varepsilon(\sigma)^2+\beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(\sigma))\,d\sigma$$
$$\leq 2K_{\alpha,\beta}\left[\varepsilon\sup_{t\in[0,T]}u^\varepsilon(t)^2+\sup_{t\in[0,T]}\varepsilon^{\eta+1}c_2^\varepsilon(t)\right]h.$$

Then, since $\eta\geq 0$ and remarking that $\varepsilon c_2^\varepsilon$ is obviously bounded by the L^1 norm of f^ε, we can use the moment estimates in Eqs. (4.3) and (4.4), so that for ε sufficiently small, there exists K independent of t and ε such that

$$\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon} x\,dx\right)\int_t^{t+s}(\alpha^\varepsilon u^\varepsilon(\sigma)^2+\beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(\sigma))\,d\sigma\leq Kh. \tag{4.16}$$

Let us now focus on the second term on the right-hand side of Eq. (4.15). Using hypotheses (H3)-(H4) and the moment estimates in Eq. (4.3), we get

$$\int_t^{t+s}\int_0^{+\infty}|a^\varepsilon(x)u^\varepsilon(\sigma)f^\varepsilon(\sigma,x)-b^\varepsilon(x)f^\varepsilon(\sigma,x)|\,dx\,d\sigma$$
$$\leq\left(K_a\sup_{\varepsilon>0}\sup_{t\in[0,T]}u^\varepsilon(t)+K_b\right)\int_t^{t+s}\int_0^{+\infty}f^\varepsilon(\sigma,x)(1+x)\,dxd\sigma.$$

Hence, there is a constant $K>0$ such that

$$\int_t^{t+s}\int_0^{+\infty}|a^\varepsilon(x)u^\varepsilon(\sigma)f^\varepsilon(\sigma,x)-b^\varepsilon(x)f^\varepsilon(\sigma,x)|\,dx\,d\sigma$$
$$\leq hK\left(\sup_{\varepsilon>0}\sup_{t\in[0,T]}\int_0^{+\infty}(1+x)f^\varepsilon(t,x)\,dx\right). \tag{4.17}$$

Combining both inequalities (4.16)-(4.17), it follows that for all $\delta>0$, for all $h\in(0,T)$,

$$\sup_{\varepsilon>0}\sup_{t\in[0,T-h]}\sup_{s\in[0,h]}\left|\int_0^{+\infty}[f^\varepsilon(t+s,x)-f^\varepsilon(t,x)]x\,dx\right|\leq\delta,$$

which gives the equicontinuity property for $\{u^\varepsilon\}$. □

The next lemma is a classical fact in the scaling used.

Lemma 3. *Let $T>0$. Let $\{\varepsilon_n\}$ a sequence converging to 0. The sequence $\{f^{\varepsilon_n}\}$ is equicontinuous in $\mathcal{M}_f((0,+\infty))$ equipped with the weak$-*$ topology.*

Proof.

By Eq. (2.1), satisfied by f^ε, if $\varphi\in\mathcal{C}^1((0,+\infty)$ has support in $[\delta,R]$, for $\varepsilon<2\delta/5$ we have $\int_{\Lambda_2^\varepsilon}\varphi(x)dx=0$, thus

$$\int_0^{+\infty}(f^\varepsilon(t+h,x)-f(t))\varphi(x)\,dx=$$
$$+\int_t^{t+h}\int_0^{+\infty}[a^\varepsilon(x)u^\varepsilon(s)f^\varepsilon(s,x)\Delta_\varepsilon\varphi(x)-b^\varepsilon(x)f^\varepsilon(s,x)\Delta_{-\varepsilon}\varphi(x)]\,dx\,ds,$$

By bounds obtained in Lemma 1 we argue as in the proof of Theorem 2.3 in [9], or see the proof of the next Lemma to conclude on the equicontinuity. □

We point out that, in the above Lemma, f^ε is seen as a measure on the open interval $(0,+\infty)$. The next lemma improves the equicontinuity of $\{f^\varepsilon\}$ around $x=0$.

LEMMA 4. *Assume $\eta \geq r_a$ and $T>0$. Let $\{\varepsilon_n\}$ a sequence converging to 0 such that $\{u^{\varepsilon_n}\}$ converges toward u uniformly on $[0,T]$ satisfying $\inf_{t\in[0,T]} u(t) > \rho$. The sequence $\{f^{\varepsilon_n}\}$ is equicontinuous in $\mathcal{M}_f([0,+\infty))$ equipped with the weak$-*$ topology.*

Proof. Let us fix $T>0$. Let $h \geq 0 \in (0,T)$, $t\in[0,T-h]$ and $s\in[0,h]$ we have, for all $\psi \in \mathcal{C}_c^\infty([0,+\infty))$ and $\varepsilon>0$

$$\begin{aligned}\left|\int_0^{+\infty} [f^\varepsilon(t+s,x)-f^\varepsilon(t,x)]\psi(x)\,dx\right| \\ \leq \int_t^{t+s} (\alpha^\varepsilon u^\varepsilon(\sigma)^2+\beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(\sigma))\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon}|\psi(x)|\,dx\right)d\sigma \\ + \int_t^{t+s}\int_0^{+\infty} |a^\varepsilon(x)u^\varepsilon(\sigma)f^\varepsilon(\sigma,x)\Delta_\varepsilon\psi(x)-b^\varepsilon(x)f^\varepsilon(\sigma,x)\Delta_{-\varepsilon}\psi(x)|\,dx\,d\sigma. \quad (4.18)\end{aligned}$$

The first integral in the right-hand side can be bounded as the following:

$$\begin{aligned}\int_t^{t+s} (\alpha^\varepsilon u^\varepsilon(\sigma)^2+\beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(\sigma))\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon}|\psi(x)|\,dx\right)d\sigma \\ \leq h\|\psi\|_\infty \sup_{t\in[0,T]}\left[\alpha^\varepsilon u^\varepsilon(t)^2+\beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(t)\right].\end{aligned}$$

Using Eqs. (4.1), (4.4) and by Proposition 2, Eq. (4.11), both terms in the supremum are uniformly bounded in time and along $\{\varepsilon_n\}$. Hence, there exists K independent of T and ε such that, for all $t\leq T-h$, $s\in[0,h]$,

$$\int_t^{t+s} (\alpha^{\varepsilon_n} u^{\varepsilon_n}(\sigma)^2+\beta^{\varepsilon_n}{\varepsilon_n}^\eta c_2^{\varepsilon_n}(\sigma))\left(\frac{1}{\varepsilon_n}\int_{\Lambda_2^{\varepsilon_n}}|\psi(x)|\,dx\right)d\sigma \leq K\|\psi|_\infty h. \quad (4.19)$$

We now focus on the second integral in the right hand side of (4.18). Using upper bounds (4.2) and (4.4), we can find a constant K such that for all $\varepsilon>0$

$$\begin{aligned}\int_t^{t+s}\int_0^{+\infty} |a^\varepsilon(x)u^\varepsilon(\sigma)f^\varepsilon(\sigma,x)\Delta_\varepsilon\psi(x)-b^\varepsilon(x)f^\varepsilon(\sigma,x)\Delta_{-\varepsilon}\psi(x)|\,dx\,d\sigma \\ \leq K\|\psi'\|_\infty\int_t^{t+s}\int_0^{+\infty} f^\varepsilon(\sigma,x)(1+x)\,dx d\sigma.\end{aligned}$$

By combining this last inequality with the moment estimate (4.3) and the inequality (4.19), there exists a constant K (not depending on ψ, h and ε), such that for all $h\in(0,T)$, $t\in[0,T-h]$, $s\in[0,h]$, $\psi\in\mathcal{C}_c^\infty([0,+\infty))$ and $n\geq 0$

$$\left|\int_0^{+\infty}[f^{\varepsilon_n}(t+s,x)-f^{\varepsilon_n}(t,x)]\psi(x)\,dx\right| \leq K(\|\psi\|_\infty+\|\psi'\|_\infty)h.$$

Let $\{\varphi_i\}_{i\geq 1}\subset\mathcal{C}_c^\infty([0,+\infty))$ be a dense subset of $\mathcal{C}_c([0,+\infty))$ for the uniform norm. The metric d defined by, for all μ and ν belonging to $\mathcal{M}_f([0,+\infty))$,

$$d(\mu,\nu)=\sum_i \frac{2^{-i}}{\|\varphi_i\|_\infty+\|\varphi_i'\|_\infty}\left|\int_0^\infty \varphi_i\mu-\int_0^\infty \varphi_i\nu\right|,$$

is equivalent to the *weak*$-*$ topology (on bounded subset), see for instance similar construction in [5, Theorem III.25]. Thus, for all $h\geq 0\in(0,T)$, we have

$$\sup_{t\in[0,T-h]}\sup_{s\in[0,h]}\sup_{n\geq 0} d(f^{\varepsilon_n}(t+s),f^{\varepsilon_n}(t))\leq Kh.$$

This concludes the proof. □

4.4. Compactness and limit

Here we give some technical lemmas which prepare the proof of the main results.

LEMMA 5. *For all $T>0$ and all $\varphi\in\mathcal{C}_c^1([0,T)\times[0,+\infty))$, we have, for all $\varepsilon>0$,*

$$\int_0^T\int_0^{+\infty}[\partial_t\varphi(t,x)+a^\varepsilon(x)u^\varepsilon(s)\Delta_\varepsilon\varphi(t,x)-b^\varepsilon(x)\Delta_{-\varepsilon}\varphi(t,x)]f^\varepsilon(t,x)\,dx\,dt$$
$$+\int_0^{+\infty}f^{in,\varepsilon}(x)\varphi(0,x)\,dx+\int_0^T[\alpha^\varepsilon u^\varepsilon(t)^2-\beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(t)]\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon}\varphi(t,x)\,dx\right)dt=0,\quad(4.20)$$

where $\Delta_h\varphi(t,x)=(\varphi(t,x+h)-\varphi(t,x))/h$, for $h\in\mathbb{R}$, and

$$u^\varepsilon(t)+\int_0^\infty xf^\varepsilon(t,x)\,dx=m^\varepsilon.\quad(4.21)$$

Proof. The proof is based on multiplying each equation of the Becker-Döring system 1.1 by $\varphi_i=\int_{\Lambda_i^\varepsilon}\varphi(t,x)\,dx$ for $\varphi\in\mathcal{C}_c^1([0,T)\times[0,+\infty))$ and using the definition of f^ε in Eq. (1.6). It is similar to Proposition 1. □

LEMMA 6. *Let $T>0$. The family $\{f^\varepsilon\}$ is relatively weak$-*$ compact in $L^\infty(0,T;w-*-\mathcal{M}_f([0,+\infty))$. If μ is an accumulation point of $\{f^\varepsilon\}$, then the restriction of μ to $(0,+\infty)$ belongs to $C([0,T],w-*-\mathcal{M}_f((0,+\infty)$, there exists a sequence $\{\varepsilon_n\}$ converging to 0 and a non-negative function $u\in C([0,T])$ such that u^{ε_n} converges to u uniformly on $[0,T]$, with $u(0)=u^{\text{in}}$ and*

$$u(t)+\int_0^\infty x\mu_t(dx)=m.$$

Moreover, for all $\varphi\in\mathcal{C}_c^1([0,T)\times[0,+\infty))$

$$\int_0^T\int_0^{+\infty}[\partial_t\varphi(t,x)+a^{\varepsilon_n}(x)u^{\varepsilon_n}(s)\Delta_{\varepsilon_n}\varphi(t,x)-b^{\varepsilon_n}(x)\Delta_{-\varepsilon_n}\varphi(t,x)]f^{\varepsilon_n}(t,x)\,dx\,dt$$
$$\to\int_0^T\int_0^{+\infty}[\partial_t\varphi(t,x)+(a(x)u(s)-b(x))\partial_x\varphi(t,x)]\mu_t(dx)\,dx$$

$$\int_0^T\alpha^{\varepsilon_n}u^{\varepsilon_n}(t)^2\left(\frac{1}{\varepsilon_n}\int_{\Lambda_2^{\varepsilon_n}}\varphi(t,x)\,dx\right)dt\to\int_0^T\alpha u(t)^2\varphi(t,0)dt,$$

and

$$\int_0^{+\infty} \varphi(0,x) f^{in,\varepsilon_n}(x)\,dx \to \int_0^{+\infty} \varphi(0,x)\,\mu^{\text{in}}(dx)$$

as $n \to +\infty$.

Proof. First, we remark that the bound against 1 in (4.3) yields to the relative compactness in $L^\infty([0,T]; w-*-\mathcal{M}_f([0,+\infty))$. Let μ an accumulation point. By Lemma 2 and bound (4.4) with Arzelá-Ascoli Theorem, entails there exists a sequence $\{\varepsilon_n\}$ and $u \in \mathcal{C}([0,T])$ such that u^{ε_n} converge to u uniformly on $[0,T]$ and $\{f^{\varepsilon_n}\}$ to μ. It remains to note that for any $\psi^\varepsilon \in \mathcal{C}_c([0,T) \times [0,+\infty))$ which converges uniformly to some ψ, we have

$$\int_0^T \int_0^\infty \psi^{\varepsilon_n}(t,x) f^{\varepsilon_n}(t,x)\,dxdt \to \int_0^T \int_0^\infty \psi(t,x)\mu_t(dx)dt,$$

as $n \to \infty$, to obtain the desired limit, see also [9, 20]. Moreover Lemma 3 improve the regularity to the continuity in time onto the space $\mathcal{M}_f((0,+\infty))$ for the *weak*$-*$ topology (open in $x=0$). Such result has been obtained for instance in [9]. Finally we obtain Eq. (4.21), using the bound (4.3) with Φ, and after regularization of the identity function, we have for all $t \in [0,T]$

$$\int_0^\infty x f^{\varepsilon_n}(t,x)\,dx \to \int_0^\infty x\mu_t(dx).$$

See [9, Proof of Theorem 2.3] for details. □

LEMMA 7. *Assume* $\mu \in L^\infty([0,T]; w-*-\mathcal{M}_f([0,+\infty))$ *is an accumulation point of* f^ε, *as considered in Lemma 6, and* u *an accumulation point of* u^ε. *Then, the limit* u *satisfies*

$$\inf_{t\in[0,T]} u(t) > \rho.$$

Proof.

If μ is an accumulation point, there exists a sequence $\{\varepsilon_n\}$ such that μ^{ε_n} converge to μ. Then, if $\varphi \in \mathcal{C}([0,T) \times (0.+\infty))$, by Lemma 5 and Lemma 6 we have that the limit satisfies (without boundary term)

$$\int_0^T \int_0^\infty \big[\partial_t \varphi(t,x) + (a(x)u(t) - b(x))\partial_x \varphi(t,x)\big]\mu(t,dx)\,dt + \int_0^\infty \varphi(0,x)\mu^{\text{in}}(dx)\,ds = 0, \quad (4.22)$$

By hypotheses on a and b and the control of $\int_0^\infty x\mu_t(dx)$ we have that

$$M_a(t) := \int_0^\infty a(x)\mu_t(dx) \text{ and } M_b(t) := \int_0^\infty b(x)\mu_t(dx).$$

Both M_a and M_b belong to $L^\infty(0,T)$. Thus, we may take $\varphi(t,x) = x\psi(t)$ in (4.22) where $\psi \in \mathcal{C}_c([0,T))$, which leads to

$$-\int_0^T \psi'(t) \int_0^\infty x\mu_t(dx)dt = \varphi(0) \int_0^\infty x\mu^{\text{in}}(dx) + \int_0^T \psi(t)u(t)M_a(t)dt - \int_0^T \psi(t)M_b(t)dt, \quad (4.23)$$

We know u is continuous on $[0,T]$ and by Eq. (4.23) *a.e.* $t\in(0,T)$

$$\frac{du(t)}{dt}=-u(t)M_a(t)+M_b(t),$$

with $u(0)=u^{\text{in}}>\rho$. It follows that

$$\frac{d}{dt}\left([u(t)-\rho]e^{\int_0^t M_a(s)ds}\right)=(M_b(t)-\rho M_a(t))e^{\int_0^t M_a(s)ds}.$$

We distinguish now two cases. First, if $r_a<r_b$, then $\rho=0$. If there exists $\tau\in(0,T)$ such that $u(\tau)=0$ and $u(t)>0$ for all $t\in[0,\tau)$, then

$$-u(0)=\int_0^\tau M_b(t)e^{\int_0^t M_a(s)ds}dt.$$

But the integral in the r.h.s is non-negative, thus $u(0)\le 0$ which contradicts $u(0)>0$. Second, if $r_a=r_b$, then $\rho>0$. If there exists $\tau\in(0,T)$ such that $u(\tau)=\rho$ and $u(t)>\rho$ for all $t\in[0,\tau)$, then,

$$-(u(0)-\rho)=\int_0^\tau (M_b(t)-\rho M_a(t))e^{\int_0^t M_a(s)ds}dt.$$

But, a.e $t\in(0,T)$

$$\int_0^\infty (b(x)-\rho a(x))\mu_t(dx)\ge 0,$$

since we assumed in this case that $a(x)\rho-b(x)\le 0$, see Eq. (2.4) in Assumption 3. This contradicts again that $u(0)>\rho$. □

Lemma 8. *Let $T>0$. Assume $\eta\ge r_a$. The family $\{f^\varepsilon\}$ is relatively compact in $\mathcal{C}([0,T];w-*-\mathcal{M}_f([0,+\infty))$. Moreover, for any sequence $\{\varepsilon_n\}$, we can extract a sub-sequence $\{\varepsilon_n'\}$ such that $\{f^{\varepsilon_n'}\}$ converges to μ with $\inf_{t\in[0,T]}u(t)>\rho$.*

Proof. Let $\{\varepsilon_n\}$ be a sequence converging to 0. By Lemmas 6 and 7, it exists a sub-sequence $\{\varepsilon_n'\}$ such that $u^{\varepsilon_n'}$ converges uniformly on $[0,T]$ to $u\in\mathcal{C}([0,T])$ such that $\inf_{t\in[0,T]}u(t)>\rho$. We may apply Lemma 4 so that $\{f^{\varepsilon_n'}\}$ is equicontinuous in $\mathcal{M}_f([0,+\infty))$. By the bound (4.3) (against 1), we have for each $t\in[0,T]$ that $\{f^{\varepsilon_n}(t):\varepsilon_n'>0\}$ belongs to a *weak*$-*$* compact set of $\mathcal{M}_f([0,+\infty))$. Thus, by Arzelá-Ascoli Theorem, the sequence $\{f^{\varepsilon_n'}\}$ is relatively compact in $\mathcal{C}([0,T];w-*-\mathcal{M}_f([0,+\infty))$. Up to a second extraction the sequence $\{f^{\varepsilon_n}\}$ admits a sub-sequence converging in $\mathcal{C}([0,T];w-*-\mathcal{M}_f([0,+\infty))$. □

5. Identification of the boundary term

This section is committed to the proof of Theorems 1 to 3. In view of Lemmas 5 to 8 it remains to identify the limit of $\varepsilon^\eta c_2^\varepsilon$ so that we can pass to the limit in the term

$$\int_0^T \beta^\varepsilon\varepsilon^\eta c_2^\varepsilon(t)\left(\frac{1}{\varepsilon}\int_{\Lambda_2^\varepsilon}\varphi(t,x)\,dx\right)dt$$

arising in (4.20).

We have separated the following in 3 subsections corresponding to the 3 theorems. Thanks to Proposition 2, the compactness of the term $\varepsilon^\eta c_2^\varepsilon$ has been already obtained in $w-*-L^\infty(0,T)$ for the first two cases, that are $\eta>r_a$ and $\eta=r_a$, and in $\mathcal{M}_f([0,T])$ by Eq. (4.5) for $\eta<r_a$. The identification of the limit relies on arguments similar to the Fenichel-Tikhonov theory on singularly perturbed dynamical systems [17]. Multiplying the re-scaled BD equations (1.1) by ε, at least formally, we have for all $t>0$ and $i\geq 2$,

$$\lim_{\varepsilon\to 0}\varepsilon\frac{d}{dt}c_i^\varepsilon=\lim_{\varepsilon\to 0}(J_{i-1}^\varepsilon(t)-J_i^\varepsilon(t))=0\,.$$

Hence, at each time $t>0$, the underlying BD model for the discrete sizes $i\geq 2$ has to reach instantaneously the equilibrium of the BD model with a constant monomer concentration $u=u(t)$. Such version of the BD model has been well studied in [26,33].

5.1. Proof of Theorem 1 – The slow de-nucleation case

Let $T>0$ and $\{\varepsilon_n\}$ a sequence converging to 0. By Lemma 8, there exists a sub-sequence, still denoted by $\{\varepsilon_n\}$ for simplicity, $\mu\in\mathcal{C}([0,T];w-*-\mathcal{M}_f([0,+\infty)))$ and $u\in\mathcal{C}([0,T])$ with $\inf_{t\in[0,T]}u(t)>\rho$ such that $\{f^{\varepsilon_n}\}$ converges to μ in $\mathcal{C}([0,T];w-*-\mathcal{M}_f([0,+\infty)))$ and u^{ε_n} converges to u uniformly on $[0,T]$. Now, applying Proposition 2, we get

$$\sup_{t\in[0,T]}\varepsilon_n^\eta c_2^{\varepsilon_n}(t)=\varepsilon_n^{\eta-r_a}\sup_{t\in[0,T]}\varepsilon^{r_a}c_2^{\varepsilon_n}(t)\to 0\,,$$

since $\eta>r_a$. Thus, combining this result with Lemma 6 we can pass to the limit in (4.20) to obtain Eq. (3.2) with $N(u)=\alpha u^2$, and Theorem 1 is proved.

5.2. Proof of Theorem 2 – The compensated nucleation case

Let $T>0$ and $\{\varepsilon_n\}$ a sequence converging to 0. We proceed similarly as above with Lemma 8 and Proposition 2. As for all $i\geq 2$, $d_i^{\varepsilon_n}=\varepsilon_n^{r_a}c_i^{\varepsilon_n}$ satisfies $d_i^{\varepsilon_n}e^{-iz}\leq F^{\varepsilon_n}(t,z)$, thanks to the estimate (4.10), there exists $z>0$ such that

$$\sup_{n\geq 0}\sup_{t\in[0,T]}\sup_{i\geq 2}d_i^{\varepsilon_n}e^{-iz}<+\infty\,.$$

Hence, by a Cantor diagonal process, we can extract another sub-sequence, still denoted by $\{\varepsilon_n\}$, such that for all $i\geq 2$,

$$d_i^{\varepsilon_n}\rightharpoonup d_i\,,\quad w-*-L^\infty(0,T)\,,$$

and

$$0\leq\sup_{t\in[0,T]}\sup_{i\geq 2}d_i(t)e^{-iz}<K_z\,. \tag{5.1}$$

We recall, from the re-scaled BD system (1.1), that the sequence $(d_i^{\varepsilon_n})_{i\geq 2}$ satisfies for each $n\geq 0$ Eq. (4.9). Hence, for all $\varphi\in\mathcal{C}^1([0,T])$,

$$\begin{aligned}{\varepsilon_n}^{1-r_a}d_i^{\varepsilon_n}(t)\varphi(t)-{\varepsilon_n}^{1-r_a}d_i^{in,\varepsilon_n}\varphi(0)-{\varepsilon_n}^{1-r_a}\int_0^t d_i^{\varepsilon_n}(s)\varphi'(s)\,ds&\\ =\int_0^t\varphi(s)\left[H_{i-1}^{\varepsilon_n}(s)-H_i^{\varepsilon_n}(s)\right]ds\,.&\end{aligned}\tag{5.2}$$

As $r_a<1$, passing to the limit $\varepsilon_n\to 0$, the left hand-side in Eq. (5.2) vanishes, and, with Assumption 3 on the kinetic rates, we have, for all $\varphi\in\mathcal{C}^1([0,T])$,

$$\int_0^T \varphi(t)\,[H_{i-1}(t)-H_i(t)]\,ds=0,$$

where $H_1=\alpha u(t)^2-\beta d_2$, and for each $i\geq 2$,

$$H_i=\begin{cases}\bar{a}i^\eta u d_i, & \text{if } \eta=r_a<r_b,\\ \bar{a}i^\eta u d_i-\bar{b}(i+1)^\eta d_{i+1}, & \text{if } \eta=r_a=r_b.\end{cases}$$

Thus, for all $i\geq 2$, we have *a.e.* $t\in(0,T)$ that $H_i(t)=H_1(t)$. In the sequel, we will distinguish two cases, $r_a<r_b$ and $r_a=r_b$.

5.2.1. The case $\eta=r_a<r_b$. In this case, $H_1=H_2$ for *a.e.* $t\in(0,T)$ yields

$$d_2(t)=\frac{\alpha u^2(t)}{\bar{a}2^\eta u(t)+\beta}.$$

Hence, the limit d_2 is uniquely identified (and by recurrence, all d_i, $i\geq 2$, using $H_i=H_1$) as a function of the limit u. Thus, combining this result with Lemma 6 we can pass to the limit in (4.20) to obtain Eq. (3.2) with $N(u)=\alpha u^2\frac{u}{u+\beta/(\bar{a}2^\eta)}$, and the case $r_a<r_b$ in Theorem 2 is proved.

5.2.2. The case $\eta=r_a=r_b$. In this case, the limit $(d_i)_{i\geq 2}$ must satisfy $H_i\equiv H$, $i\geq 1$, for a given constant H. We classically (in the study of the equilibrium states of BD equations [1]) define $Q_1=1$ and for all $i\geq 2$,

$$Q_i=\frac{\alpha}{\beta}\prod_{k=2}^{i-1}\frac{\bar{a}k^{r_a}}{\bar{b}(k+1)^{r_a}},\ i\geq 2.$$

The solutions that satisfy $H_i\equiv H$ for all $i\geq 1$, are given by, after some algebraic manipulation (see [26, lemma 1]),

$$d_i=Q_iu^i\Big(1-H\frac{1}{\alpha u^2}-H\sum_{k=2}^{i-1}\frac{1}{\bar{a}k^{r_a}Q_ku^{k+1}}\Big),\quad i\geq 2.$$

Thus, for all $i\geq 2$,

$$d_i=\frac{\alpha u^2}{\beta}\frac{2^{r_a}}{i^{r_a}}\Big(\frac{\bar{a}u}{\bar{b}}\Big)^{i-2}\left[1-\frac{H}{\alpha u^2}\left(1+\frac{\beta}{2^{r_a}}\frac{1}{\bar{a}u-\bar{b}}\right)+\frac{H\beta}{\alpha u^2 2^{r_a}}\frac{\big(\bar{b}/(\bar{a}u)\big)^{i-2}}{\bar{a}u-\bar{b}}\right].$$

However, for $u(t)>\rho=\bar{b}/\bar{a}$, there exists a unique H such that the bound (5.1) is satisfied, given by

$$H=\frac{\alpha u^2}{\Big(1+\frac{\beta}{2^\eta}\frac{1}{\bar{a}u-\bar{b}}\Big)}=\frac{\alpha u^2(\bar{a}u-\bar{b})}{\bar{a}u+\frac{\beta}{2^\eta}-\bar{b}}.$$

For this value, we have *a.e.* $t\in[0,T]$

$$d_2(t)=\frac{\alpha u(t)^2}{2^\eta(\bar{a}u-\bar{b})+\beta}=\frac{\alpha u(t)^2}{\beta}\left[1-\frac{\bar{a}u-\bar{b}}{\bar{a}u-\bar{b}+\beta/2^\eta}\right].$$

Hence, proceeding as before we recover the second part of Theorem 2.

5.3. Proof of Theorem 3 – The fast de-nucleation

In the case $\eta < r_a$ we have no L^∞ bound over $\varepsilon^\eta c_2^\varepsilon$, and no equicontinuity property on $\{f^\varepsilon\}$ in $\mathcal{M}_f([0,+\infty))$. Nevertheless, we can apply Lemma 6 and 7 . Thus, let $T>0$ and $\{\varepsilon_n\}$ a sequence converging to 0, there exists a sub-sequence of $\{\varepsilon_n\}$ (not re-labeled), $\mu \in L^\infty([0,T]; w-*-\mathcal{M}_f([0,+\infty)))$ and $u \in \mathcal{C}([0,T])$ such that $f^{\varepsilon_n} \rightharpoonup \mu$ in $w-*-L^\infty([0,T]; w-*-\mathcal{M}_f([0,+\infty)))$ and u^{ε_n} converges uniformly to u on $[0,T]$ such that $\inf_{t\in[0,T]} u(t) > \rho$. Note, Lemma 7 also provides the regularity in time of the restriction to $(0,+\infty)$ of the limit. Moreover, by the bound (4.5) we can extract another sub-sequence of $\{\varepsilon_n\}$ (not re-labeled) such that $d_2^{\varepsilon_n} := \varepsilon_n^\eta c_2^\varepsilon$ converges to a non-negative finite measure Γ_2 on $[0,T]$, where the convergence holds in $\mathcal{M}_f([0,T])$ endowed with the $weak-*$ topology. Also, for all $\varphi \in \mathcal{C}^1([0,T])$, the equation (1.1) for $i=2$ yields

$$\begin{aligned}
{\varepsilon_n}^{1-r_a}{\varepsilon_n}^{r_a} c_2^{\varepsilon_n}(T)\varphi(T) - {\varepsilon_n}^{1-r_a}{\varepsilon_n}^{r_a} c_2^{in,\varepsilon_n}\varphi(0) - {\varepsilon_n}^{1-r_a}\int_0^T \varphi'(t){\varepsilon_n}^{r_a} c_2^{\varepsilon_n}(t)dt \\
= \int_0^T \varphi(t)[\alpha^{\varepsilon_n} u^{\varepsilon_n}(t)^2 - \beta^{\varepsilon_n} d_2^{\varepsilon_n}(t)]dt \\
- \int_0^T \varphi(t)[\overline{a}_2^{\varepsilon_n}{\varepsilon_n}^{r_a-\eta} u^{\varepsilon_n}(t) d_2^{\varepsilon_n}(t) - \overline{b}_3^{\varepsilon_n}{\varepsilon_n}^{r_b} c_3^{\varepsilon_n}(t)]dt. \quad (5.3)
\end{aligned}$$

By Proposition 2, ${\varepsilon_n}^{r_a} c_2^{\varepsilon_n}(t)$ is uniformly bounded with respect to both time $t \in [0,T]$ and n, so that the left hand side of Eq. (5.3) goes to 0 as $\varepsilon_n \to 0$. Hence, with the bound (4.5) and since $\eta < r_a$, we have

$$\lim_{\varepsilon_n \to 0} \int_0^T \varphi(t){\varepsilon_n}^{r_b} c_3^{\varepsilon_n}(t)dt = \frac{1}{\overline{b}_3}\left(\int_0^T \varphi(t)\beta\Gamma_2(dt) - \int_0^T \varphi(t)\alpha u(t)^2 dt\right). \quad (5.4)$$

Here again, two cases have to be considered, $r_a < r_b$ and $r_a = r_b$.

5.3.1. The case $r_a < r_b$. In this case, we use again Proposition 2 for the left hand-side of Eq. (5.4), and use that $\varepsilon^{r_b - r_a} \to 0$ as $\varepsilon_n \to 0$. Thus, we are led with the following equality in measure

$$\Gamma_2(dt) = \frac{\alpha}{\beta} u(t)^2 dt.$$

Thus, combining this result with Lemma 6 we can pass to the limit in (4.20) and we obtain the first case of Theorem 3.

5.3.2. The case $r_a = r_b$. In this case, we use again the fact that by Proposition 2, up to a sub-sequence of $\{\varepsilon_n\}$ (not relabeled), for all $i \geq 2$, there exists $d_i \in L^\infty(0,T)$ and $z_0 > 0$ such that

$${\varepsilon_n}^{r_b} c_i^{\varepsilon_n} \rightharpoonup d_i \quad w-*-L^\infty(0,T),$$

and for all $z < z_0$, there exists $K_z > 0$ such that

$$0 \leq \sup_{t\in[0,T]} \sup_{i\geq 2} d_i(t) e^{-iz} < K_z. \quad (5.5)$$

From Eq. (5.4), we obtain the equality in measure

$$\overline{b}_3 d_3\, dt = \beta\Gamma_2(dt) - \alpha u(t)^2\, dt.$$

Then, iterating the procedure, from equation (1.1), we get that, for all $i \geq 3$ and $\varphi \in \mathcal{C}^1([0,T])$

$$\begin{aligned}
{\varepsilon_n}^{1-r_a}{\varepsilon_n}^{r_a}c_i^{\varepsilon_n}(T)\varphi(T) - {\varepsilon_n}^{1-r_a}{\varepsilon_n}^{r_a}c_i^{in,\varepsilon_n}\varphi(0) - {\varepsilon_n}^{1-r_a}\int_0^T \varphi'(t){\varepsilon_n}^{r_a}c_i^{\varepsilon_n}(t)dt \\
= \int_0^T \varphi(t)[\overline{a}_{i-1}^{\varepsilon_n}u^{\varepsilon_n}(t){\varepsilon_n}^{r_a}c_{i-1}^{\varepsilon_n}(t) - \overline{b}_i^{\varepsilon_n}{\varepsilon_n}^{r_a}c_i^{\varepsilon_n}(t)]\,dt \\
- \int_0^T \varphi(t)[\overline{a}_i^{\varepsilon_n}u^{\varepsilon_n}(t){\varepsilon_n}^{r_a}c_i^{\varepsilon_n}(t) - \overline{b}_{i+1}^{\varepsilon_n}{\varepsilon_n}^{r_a}c_{i+1}^{\varepsilon_n}(t)]dt.
\end{aligned}$$

Hence, for $i=3$, writing ${\varepsilon_n}^{r_a}c_2^{\varepsilon_n}(t) = \varepsilon_n^{r_a-\eta}d_2^{\varepsilon_n}(t) \to 0$ (in $\mathcal{M}_f([0,T])$), we obtain

$$0 = \int_0^T \varphi(t)[-\overline{b}_3 d_3(t) - \overline{a}_3 u(t) d_3(t) + \overline{b}_4 d_4(t)]dt.$$

And for all $i \geq 4$,

$$0 = \int_0^T \varphi(t)[\overline{a}_{i-1}u(t)d_{i-1}(t) - \overline{b}_i d_i(t) - \overline{a}_i u(t) d_i(t) + \overline{b}_{i+1}d_{i+1}(t)]dt.$$

With $H_2 = -\overline{b}_3 d_3$, $H_i = \overline{a}_i u^{\varepsilon} d_i(t) - \overline{b}_{i+1}d_{i+1}$, $i \geq 3$, then we must have a.e. $H_i = H_2 =: H$, for all $i \geq 2$. Then we get, for all $i \geq 3$,

$$d_i(t) = -\frac{H}{\overline{b}_i}\sum_{j=3}^{i}\left(\prod_{k=j}^{i-1}\frac{\overline{a}_k}{\overline{b}_k}\right)u^{(i-j)} = -\frac{H}{\overline{b}_i}\sum_{j=3}^{i}\left(\frac{\overline{a}u}{\overline{b}}\right)^{i-j}.$$

In order to fulfill the bound (5.5), we must get $H=0$, so that $d_3=0$ and the following equality in measure holds

$$\Gamma_2(dt) = \frac{\alpha}{\beta}u(t)^2 dt.$$

This ends the proof of Theorem 3.

6. Extension to a density

In this section, we make an extra-assumption in order to obtain a convergence result in L^1 functional space, so that the limit measure has a density with respect to the Lebesgue measure:

Assumption 5. *There is $\delta \in (0, 1/r_a - 1)$ such that, for the function $\Psi(y) = y^{1+\delta}$,*

$$\sup_{\varepsilon>0}\int_0^{\infty}\Psi(f^{in,\varepsilon}(x))dx < \infty. \tag{H8}$$

Moreover, the kinetic rates are given by exact power law functions, i.e.,

$$\begin{aligned}
a_i^{\varepsilon} &= \overline{a}(\varepsilon i)^{r_a}, \; i \geq 2, \\
b_i^{\varepsilon} &= \overline{b}(\varepsilon i)^{r_b}, \; i \geq 3.
\end{aligned} \tag{H9}$$

Remark 16. *The first hypothesis* (H8) *is slightly stronger than a compactness hypothesis in $L^1(dx)$, where a more general* (*and not explicit*) *Ψ can be obtained, see* [7].

However, having an explicit power law function for Ψ will simplify the following calculus. The same is valid for the extra hypothesis (H9) *on the kinetic rates, which is in agreement with hypothesis* (H5).

Assuming assumptions 1-5 hold true, we can now prove the last result.

THEOREM 4. *Assume $\eta \geq r_a$ and $r_a = r_b$. Let $\{\varepsilon_n\}$ be a sequence converging to* 0. *There exists $T > 0$, a sub-sequence $\{\varepsilon_{n'}\}$ of $\{\varepsilon_n\}$, and $f \in \mathcal{C}([0,T], w - L^1(\mathbb{R}_+, x^{r_a\delta}dx)) \cap L^\infty([0,T]; L^1(\mathbb{R}_+, (1+x)dx))$ such that the measure $f(t,x)dx$ is a N-solution of LS with mass m and*

$$f^{\varepsilon_{n'}} \xrightarrow[n' \to +\infty]{} f$$

in $\mathcal{C}([0,T]; w - L^1(\mathbb{R}_+, x^{r_a\delta}dx))$. N is given in Theorem 1-2 according to the value of η.

The proof of this theorem is based on the following lemma which proof is postponed below

LEMMA 9. *Assume $\eta \geq r_a$ and $r_a = r_b$. Let a sequence $\{\varepsilon_n\}$ converging to* 0. *There exist $T > 0$ and a sub-sequence $\{\varepsilon_{n'}\}$ of $\{\varepsilon_n\}$ such that*

$$\sup_{n' \geq 0} \sup_{t \in [0,T]} \int_0^\infty \min(1, x^{r_a\delta}) \Psi(f^{\varepsilon_{n'}}(t,x))\, dx < +\infty.$$

Proof of Theorem 4. Using the same proof as for Theorem 1 and 2, we obtain a sub-sequence $f^{\varepsilon_{n'}}$ that converges in measure. We now remark that, combining the estimates (4.3) in Lemma 1 and the last Lemma 9 we can apply the Dunford-Pettis theorem, and we have a weak compact subset $\mathcal{K}$ of $L^1(\mathbb{R}_+, x^{r\delta}dx)$ such that for all $t \in [0,T]$ and $n' \geq 0$, $f^{\varepsilon_{n'}}(t) \in \mathcal{K}$. We are now in position to prove that along another subsequence, still denoted by $\{\varepsilon_{n'}\}$, the sequence converges to some f in $C([0,T], w - L^1(\mathbb{R}_+, x^{r\delta}dx))$. Moreover, f belongs to $L^\infty([0,T], L^1(\mathbb{R}_+, (1+x)dx)$. The proof follows similar arguments as in [20, Proof of Theorem 2.2, p. 981] which consists in proving the equicontinuity of

$$t \to \int_0^R f^\varepsilon(t,x)\varphi(x)x^{r\delta}dx,$$

for all $\varphi \in L^\infty(0,R)$ and $R > 0$. Indeed, by Eq. (4.3) we have for any $\varphi \in \mathcal{C}^1$ with compact support in $(0,R)$ that (see also the proof of lemma 4)

$$\lim_{h \to 0} \sup_{t \in [0,T-h]} \sup_{s \in (0,h)} \left| \int_0^\infty (f^\varepsilon(t+s,x) - f^\varepsilon(t,x))\varphi(x)x^{r\delta}dx \right| = 0.$$

Then taking a pointwise convergent sequence $\{\varphi^n\}$ in $\mathcal{C}_c([0,R])$ of $\varphi \in L^\infty(0,R)$ and using Egorov's theorem we get the desire result. Finally, we apply a variant of Arzela-Ascoli theorem for weak topology, see [31, Theorem 1.3.2], so that for each $R > 0$, the sequence is relatively compact in $C([0,T], w - L^1((0,R), r^{r\delta}dx)$. By the compact containment we improve this results on $\mathbb{R}_+$.

6.1. Technical results

Before proving Lemma 9, we start by some technical lemmas.

LEMMA 10. *Let $\varphi \in \mathcal{C}_b([0,\infty))$ non-negative. Then, for any $I \geq 3$,*

$$\begin{aligned}\int_0^\infty \varphi(x)\left[\Psi(f^\varepsilon(t,x)) - \Psi(f^{in,\varepsilon}(x))\right] dx \\ \leq \varepsilon \sum_{i=2}^{I-1} \varphi_i^\varepsilon \Psi(c_i^\varepsilon(t)) + \int_0^t \left[\varphi_I^\varepsilon a_{I-1}^\varepsilon u^\varepsilon(s)\Psi(c_{I-1}^\varepsilon(s)) - \varphi_{I-1}^\varepsilon b_I^\varepsilon \Psi(c_I^\varepsilon(s))\right] ds \\ + \int_0^t \int_{(I-1/2)\varepsilon}^\infty \Big[a^\varepsilon(x)u^\varepsilon(s)\Delta_\varepsilon \varphi(x) - b^\varepsilon(x)\Delta_{-\varepsilon}\varphi(x) \\ - \delta\left(u^\varepsilon(s)\Delta_{-\varepsilon}a^\varepsilon(x) - \Delta_\varepsilon b^\varepsilon(x)\varphi(x)\right)\Big]\Psi(f^\varepsilon(x,s))dxds. \quad (6.1)\end{aligned}$$

where $\varphi_i^\varepsilon = 1/\varepsilon \int_{\Lambda_i^\varepsilon} \varphi(x)\,dx$.

Proof. The proof follows similar lines as in [20, Lemma 4.1], but we take profit of the explicit form of Ψ to obtain a necessary finer estimate. We sketch it briefly below. From the (BD) system (1.1), it comes

$$\begin{aligned}\int_0^\infty \varphi(x)\left[\Psi(f^\varepsilon(t,x)) - \Psi(f^{in,\varepsilon}(x))\right] dx = \sum_{i\geq 2} \int_{\Lambda_i^\varepsilon} \varphi(x)\left[\Psi(c_i^\varepsilon(t)) - \Psi(c_i^\varepsilon(0))\right] dx \\ = \varepsilon \sum_{2\leq i\leq I-1} \varphi_i^\varepsilon \left[\Psi(c_i^\varepsilon(t)) - \Psi(c_i^\varepsilon(0))\right] + \sum_{i\geq I} \varphi_i^\varepsilon \int_0^t \left[J_{i-1}^\varepsilon(s) - J_i^\varepsilon(s)\right]\Psi'(c_i^\varepsilon(s))\,ds.\end{aligned}$$

We can decompose the latter in three parts,

$$\int_0^\infty \varphi(x)\left[\Psi(f^\varepsilon(t,x)) - \Psi(f^{in,\varepsilon}(x))\right] dx = N^\varepsilon(t) + \int_0^t \left[A^\varepsilon(s) + B^\varepsilon(s)\right] ds,$$

where

$$\begin{aligned}N^\varepsilon(t) &:= \varepsilon \sum_{2\leq i\leq I-1} \varphi_i^\varepsilon \left[\Psi(c_i^\varepsilon(t)) - \Psi(c_i^\varepsilon(0))\right], \\ A^\varepsilon(t) &:= \sum_{i\geq I} \varphi_i^\varepsilon u^\varepsilon(t)[a_{i-1}^\varepsilon c_{i-1}^\varepsilon(t) - a_i^\varepsilon c_i^\varepsilon(t)]\Psi'(c_i^\varepsilon(t)), \\ B^\varepsilon(t) &:= \sum_{i\geq I} \varphi_i^\varepsilon [b_{i+1}^\varepsilon c_{i+1}^\varepsilon(t) - b_i^\varepsilon c_i^\varepsilon(t)]\Psi'(c_i^\varepsilon(t)).\end{aligned}$$

Then, in A^ε we can re-write, using the convexity of Ψ, for all $i \geq I$,

$$\begin{aligned}[a_{i-1}^\varepsilon c_{i-1}^\varepsilon(t) - a_i^\varepsilon c_i^\varepsilon(t)]\Psi'(c_i^\varepsilon(t)) \\ = a_{i-1}^\varepsilon [c_{i-1}^\varepsilon(t) - c_i^\varepsilon(t)]\Psi'(c_i^\varepsilon(t)) + (a_{i-1}^\varepsilon - a_i^\varepsilon)c_i\Psi'(c_i^\varepsilon(t)) \\ \leq a_{i-1}^\varepsilon \left(\Psi(c_{i-1}^\varepsilon(t)) - \Psi(c_i^\varepsilon(t))\right) + (a_{i-1}^\varepsilon - a_i^\varepsilon)c_i^\varepsilon(t)\Psi'(c_i^\varepsilon(t)).\end{aligned}$$

Then, reordering the term in the last inequality and then using that $x\Psi'(x) - \Psi(x) = \delta\Psi(x)$,

$$\begin{aligned}[a_{i-1}^\varepsilon c_{i-1}^\varepsilon(t) - a_i^\varepsilon c_i^\varepsilon(t)]\Psi'(c_i^\varepsilon(t)) \\ \leq a_{i-1}^\varepsilon \Psi(c_{i-1}^\varepsilon(t)) - a_i^\varepsilon \Psi(c_i^\varepsilon(t)) + (a_{i-1}^\varepsilon - a_i^\varepsilon)[c_i^\varepsilon(t)\Psi'(c_i^\varepsilon(t)) - \Psi(c_i^\varepsilon(t))] \\ = a_{i-1}^\varepsilon \Psi(c_{i-1}^\varepsilon(t)) - a_i^\varepsilon \Psi(c_i^\varepsilon(t)) - \delta(a_i^\varepsilon - a_{i-1}^\varepsilon)\Psi(c_i^\varepsilon(t)).\end{aligned}$$

Thus, we obtain for A the following estimate,

$$A^\varepsilon(t) \le \sum_{i\ge I} a_i^\varepsilon u^\varepsilon(\varphi_{i+1}^\varepsilon - \varphi_i^\varepsilon)\Psi(c_i^\varepsilon(t)) + \varphi_I^\varepsilon a_{I-1}^\varepsilon u^\varepsilon(t)\Psi(c_{I-1}^\varepsilon(t)) \\ - \delta u^\varepsilon \sum_{i\ge I} \varphi_i^\varepsilon (a_i^\varepsilon - a_{i-1}^\varepsilon)\Psi(c_i^\varepsilon).$$

We estimate B, by similar argument, to get,

$$B^\varepsilon(t) \le \sum_{i\ge I} \varphi_i^\varepsilon [b_{i+1}^\varepsilon \Psi(c_{i+1}^\varepsilon) - b_i^\varepsilon \Psi(c_i^\varepsilon)] + \delta \sum_{i\ge I} \varphi_i^\varepsilon (b_{i+1}^\varepsilon - b_i^\varepsilon)\Psi(c_i^\varepsilon) \\ \le \sum_{i\ge I} (\varphi_{i-1}^\varepsilon - \varphi_i^\varepsilon) b_i^\varepsilon \Psi(c_i^\varepsilon) - \varphi_{I-1}^\varepsilon b_I^\varepsilon \Psi(c_I^\varepsilon) + \delta \sum_{i\ge I} \varphi_i^\varepsilon (b_{i+1}^\varepsilon - b_i^\varepsilon)\Psi(c_i^\varepsilon).$$

Both estimates on A^ε and B^ε directly give (6.1). □

LEMMA 11. *For all $0 \le r < 1$, and for all $0 < \delta < \frac{1}{r} - 1$, there exists I_0 such that for all $i \ge I_0$, and all $x \in [0,1]$,*

$$\left[i^r \left((i+1/2+x)^{r\delta} - (i-1/2+x)^{r\delta} \right) - \delta (i^r - (i-1)^r)(i-1/2+x)^{r\delta} \right] \le 0,$$

Proof. Doing an expansion as $i \to \infty$, we easily obtain

$$\left[i^r \left((i+1/2+x)^{r\delta} - (i-1/2+x)^{r\delta} \right) - \delta (i^r - (i-1)^r)(i-1/2+x)^{r\delta} \right] \\ = r\delta \frac{i^r (i - \frac{1}{2} + x)^{r\delta}}{i^2} \left[\frac{r(1+\delta)-1}{2} - x + O(\frac{1}{i}) \right].$$

We conclude straightforwardly as $r(1+\delta) - 1 < 0$. □

6.2. Proof of Lemma 9

In the following, let $r = r_a = r_b$ and $I = I_0$ given by Lemma 11. We want to bound each term of Eq. (6.1) with $\varphi(x) = \min(1, x^{r\delta})$. Remark the term $-\varphi_{I_0-1}^\varepsilon b_{I_0}^\varepsilon \Psi(c_{I_0}^\varepsilon(t))$ can be easily dropped in Eq. (6.1) since it is non-positive. Also, note that, for $2 \le i \le I_0$,

$$\varepsilon \varphi_i^\varepsilon \Psi(c_i^\varepsilon(t)) \le \varepsilon^{1-r(1+\delta)} \varphi_i^\varepsilon \left(\varepsilon^r c_i^\varepsilon(t)\right)^{1+\delta}.$$

Thus, since φ_i^ε is bounded and $\delta \le 1/r - 1$, we apply Lemma 8 and Proposition 2 to obtain $T > 0$ and a sub-sequence, still denoted by $\{\varepsilon_n\}$, to get:

$$\sup_{n\ge 0} \sup_{t\in[0,T]} \left(\varepsilon_n \varphi_i^{\varepsilon_n} \Psi(c_i^{\varepsilon_n}(t))\right) < \infty. \tag{6.2}$$

Similarly, using that $u^\varepsilon(t) \le K_m$, we have

$$\varphi_{I_0}^\varepsilon a_{I_0-1}^\varepsilon u^\varepsilon(t) \Psi(c_{I_0-1}^\varepsilon(t)) = \overline{a}(I_0-1)^r u^\varepsilon(t) \left(\int_{I_0-1/2}^{I_0+1/2} y^{r\delta} dy \right) \left(\varepsilon^r c_{I_0-1}^\varepsilon(t)\right)^{1+\delta} \\ \le K_m \overline{a}(I_0-1)^r \left(\int_{I_0-1/2}^{I_0+1/2} y^{r\delta} dy \right) \sup_{\varepsilon>0} \sup_{t\in[0,T]} \left(\varepsilon^r c_{I_0-1}^\varepsilon(t)\right)^{1+\delta} < \infty, \tag{6.3}$$

By these estimates, the boundary terms in Eq. (6.1) are uniformly bounded. We are led with the remaining integral term on $((I_0-1/2)\varepsilon,\infty)$. Denote, for all $\varepsilon>0$ and $x>0$,

$$D^\varepsilon(x)=a^\varepsilon(x)u^\varepsilon(t)\Delta_\varepsilon\varphi(x)-b^\varepsilon(x)\Delta_{-\varepsilon}\varphi(x)-\delta\left(u^\varepsilon\Delta_{-\varepsilon}a^\varepsilon(x)-\Delta_\varepsilon b^\varepsilon(x)\right)\varphi(x).$$

Thus,

$$\begin{aligned}\int_{(I_0-1/2)\varepsilon}^1 & D^\varepsilon(x)\Psi(f^\varepsilon(x,t))dx\\ &=\sum_{i=I_0}^{1/\varepsilon}\frac{1}{\varepsilon}\int_{\Lambda_i^\varepsilon}\Big[(a_i^\varepsilon u^\varepsilon(t)(\varphi(x+\varepsilon)-\varphi(x))-b_i^\varepsilon(\varphi(x)-\varphi(x-\varepsilon)))\\ &\qquad-\delta\left(u^\varepsilon(a_i^\varepsilon-a_{i-1}^\varepsilon)-(b_{i+1}^\varepsilon-b_i^\varepsilon)\right)\varphi(x)\Big]\Psi(c_i^\varepsilon(t))dx.\end{aligned}$$

Then, on $x\in(0,1)$, we have that $\varphi(x)=x^{r\delta}$, and letting $\Gamma_i=[i-1/2,i+1/2)$ and changing variable $\varepsilon y=x$, we obtain

$$\begin{aligned}\int_{(I_0-1/2)\varepsilon}^1 & D^\varepsilon(x)\Psi(f^\varepsilon(x,t))dx\\ &=\sum_{i=I_0}^{1/\varepsilon}\varepsilon^{r(1+\delta)}\int_{\Gamma_i}\Big[\left(\overline{a}i^r u^\varepsilon(t)((y+1)^{r\delta}-y^{r\delta})-\overline{b}i^r(y^{r\delta}-(y-1)^{r\delta})\right)\\ &\qquad-\delta\left(u^\varepsilon\overline{a}(i^r-(i-1)^r)-\overline{b}((i+1)^r-i^r)\right)y^{r\delta}\Big]\Psi(c_i^\varepsilon(t))dy.\end{aligned}$$

Finally, rearranging the term we have

$$\begin{aligned}\int_{(I_0-1/2)\varepsilon}^1 & D^\varepsilon(x)\Psi(f^\varepsilon(x,t))dx\\ &=\sum_{i=I_0}^{1/\varepsilon}\varepsilon^{r(1+\delta)}\int_{\Gamma_i}\Big[\left(\overline{a}u^\varepsilon(t)-\overline{b}\right)\left(i^r((y+1)^{r\delta}-y^{r\delta})-\delta(i^r-(i-1)^r)y^{r\delta}\right)\\ &\qquad+\overline{b}i^r\left((y+1)^{r\delta}-2y^{r\delta}+(y-1)^{r\delta}\right)\\ &\qquad+\delta\overline{b}((i+1)^r-2i^r+(i-1)^r)y^{r\delta}\Big]\Psi(c_i^\varepsilon(t))dy.\end{aligned}$$

Then, as the second discrete derivative are negative, that is, for all $s<1$ and all $x>1$,

$$((x+1)^s-2x^s+(x-1)^s)\le 0,$$

we obtain

$$\begin{aligned}\int_{(I_0-1/2)\varepsilon}^1 & D^\varepsilon(x)\Psi(f^\varepsilon(x,t))dx\\ &\le\varepsilon^{r(1+\delta)}\left(\overline{a}u^\varepsilon(t)-\overline{b}\right)\sum_{i=I_0}^{1/\varepsilon}\int_{\Lambda_i}\Big[i^r((y+1)^{r\delta}-y^{r\delta})\\ &\qquad-\delta(i^r-(i-1)^r)y^{r\delta}\Big]\Psi(c_i^\varepsilon(t))dy.\end{aligned}$$

The term under the integral is negative by Lemma 11. We now fix $T>0$ and extract a sub-sequence $\{\varepsilon_{n'}\}$ given by Lemma 8 such that $\overline{a}u^{\varepsilon}(t)-\overline{b}>0$ on $[0,T]$. Thus,

$$\int_{(I_0-1/2)\varepsilon}^{1} D^{\varepsilon}(x)\Psi(f^{\varepsilon}(x,t))dx \leq 0. \tag{6.4}$$

On the other hand we have, since $\Delta_{\varepsilon}\varphi=0$ on $(1,+\infty)$,

$$\int_1^{\infty}\Big[D^{\varepsilon}(x)\Psi(f^{\varepsilon}(x,t))dx \leq \delta(K_m\sup_{x\geq 1}|a'(x)|+\sup_{x\geq 1}|b'(x)|)\int_1^{\infty}\varphi(x)\Psi(f^{\varepsilon}(x,t))dx, \tag{6.5}$$

and we conclude by estimates (6.2) to (6.5) that, for some constant $K>0$ and all $t\in[0,T]$,

$$\int_0^{\infty}\varphi(x)\Psi(f^{\varepsilon_n}(t,x)) \leq K+\int_0^{\infty}\Psi(f^{in,\varepsilon_n}(x))\,dx+K\int_0^t\int_0^{\infty}\varphi(x)\Psi(f^{\varepsilon_n}(t,x)).$$

We conclude the proof with the Gronwall Lemma.

6.3. The general case

The main difficulty to face the case $r_a<r_b$ is to find a test function φ in Eq. (6.1) which make the term under the integral negative around 0, but which also keep the boundary terms bounded. We believe that a good function would be

$$\varphi(x)=\min(x^{r\delta}e^{-Kx^{r_b-r_a}},c),$$

for some $c>0$ small and $K>0$ large enough. It recovers the case $r_a=r_b$ (with $c=1$). Computations are not presented here because too fastidious. Just let us show that, at the limit $\varepsilon\to 0$,

$$[\overline{a}x^{r_a}u(t)-\overline{b}x^{r_b}]\varphi'(x)-\delta\left[r_a\overline{a}x^{r_a-1}u(t)-r_b\overline{b}x^{r_b-1}\right]\varphi(x) = \frac{\varphi(x)}{x}(r_b-r_a)\left[\delta\overline{b}x^{r_b}-Kx^{r_b-r_a}(\overline{a}x^{r_a}u(t)-\overline{b}x^{r_b})\right].$$

But since $u(t)>\rho$, it exists $x_0>0$ small and $\gamma>0$ such that the flux is bounded from below by $\overline{a}x^{r_a}u(t)-\overline{b}x^{r_b}\geq\gamma\overline{a}x^{r_a}$ on $[0,x_0]$, thus

$$[\overline{a}x^{r_a}u(t)-\overline{b}x^{r_b}]\varphi'(x)-\delta\left[r_a\overline{a}x^{r_a-1}u(t)-r_b\overline{b}x^{r_b-1}\right]\varphi(x) \leq \frac{\varphi(x)}{x}(r_b-r_a)\left[\delta\overline{b}-K\gamma\right]x^{r_b}.$$

Hence, for K large enough the term is negative around 0, which was the essential ingredient of the proof of Theorem 4.

7. Discussion

In this work, we obtained limit theorems to derive rigorously the link between a discrete-size coagulation-fragmentation model, the Becker-Döring (BD) model, and a continuous-size model, the Lifshitz-Slyozov (LS) model. We used a weak-convergence

in measure, to prove that a sequence of discrete stepwise functions associated to the BD model converges towards a measure solution of the LS model. The originality of our work, compared to previous works in [9, 20], consists of being able to rigorously define a boundary flux condition, for the limit non-linear transport partial differential equation of the LS model. This boundary condition has been obtained thanks to an averaging procedure for the smaller-sized cluster, namely the one of size $i=2$. It is classical when passing from a discrete to a continuous model (think of a random walk converging to a Brownian motion) to accelerate the rates (or equivalently, the time) between each discrete transition. Hence, each individual discrete-size cluster evolves in the re-scaled BD model (1.1) at a faster time scale than the continuous density function f^ε in Eq. (2.1). Although the fast-motion involves a dynamical system of infinite dimension, we could obtain appropriate L^∞-bounds on the time trajectories of each discrete-sized cluster, and prove that, in the limit when the scaling parameter $\varepsilon \to 0$, each discrete-sized cluster is the unique solution of an algebraic equation, which appears to be the same as the steady-state condition of a constant monomer BD model.

Let us now discuss in more details what were the scaling assumptions that led to the study of the system (1.1) (for the mathematical derivation, see the appendix 7). Roughly, the system (1.1) is obtained when we consider that the clusters have very large sizes but are present in a small quantity compared to a large excess of free particles. The re-scaled equations are obtained in a large volume hypothesis, and the scaling of the macroscopic reaction rates accounts for the volume-dependence of the aggregation (so that aggregation and fragmentation occur at the same time scale).

However, importantly enough, the first aggregation (nucleation) rate is scaled differently from the other aggregation rates (see Appendix 7) and this comes from the special role played by the free particles. Despite the large excess of free particles, in this framework, the nucleation occurs at the same time scale than the aggregation of large-sized clusters, and consequently prevents the formation of too many clusters. A different choice at this step would lead to a rapid depletion of free particles, and would result in different mass conservation where free particles are not present as a distinct entity any more– see the work [20] on the Lifshitz-Slyozov-Wagner equation.

Finally, we allowed a flexibility in the scaling of the first fragmentation (de-nucleation), quantified by the exponent η. We found (see Theorems 1-2-3) that different values of η give rise to distinct boundary conditions at the limit when ε goes to 0. The most natural case, $\eta = r_b$, corresponds to the case where the clusters of size 2 dissociate at the same speed than the small-sized clusters of size i, $i \geq 3$. Then, the case $\eta > r_b$ corresponds to an asymptotically irreversible nucleation (and leads to a macroscopic flux $N(t) = \alpha u(t)^2$, which corresponds to the microscopic nucleation rate – this conclusion actually holds for all $\eta > r_a$). And the case $\eta \leq r_a < r_b$ corresponds to a strongly reversible de-nucleation (and leads to $0 \leq N(t) < \alpha u(t)^2$ according to the value r_a).

We emphasize what this scaling means in terms of application, and in particular for the amyloid formation literature described in the introduction [2, 12, 14, 15, 21, 29] . First, the pre-equilibrium hypothesis for the small clusters was found to be valid in our framework, meaning that if each discrete-size cluster evolves at a fast time scale, their behavior can be nicely summed up by an appropriate boundary condition in a continuous-size PDE. However, due to the specific form of the BD reactions, to recover a LS equation with boundary condition, as used in [2, 15, 29], it is important to realize that the first aggregation rate, leading to the formation of clusters of size 2, cannot be too fast, and needs (in our framework) actually to be one order of magnitude slower than subsequent aggregation rates. Interestingly, we were able to find both a positive

boundary condition, similar as used in fibrils formation models [2,15,29], and a zero flux boundary condition, as used in the prion equation [12,14,21], according to the order of the fragmentation rate magnitude for the cluster of size 2, compared to the other rates. Indeed, consistently with the literature, we found that if clusters of size 2 degrade very fast into free particles ($\eta \leq r_a < r_b$), the appropriate boundary condition is a zero-flux boundary condition.

Hence, our work shed lights on which appropriate boundary conditions should be used for the LS equation (or similar continuous coagulation models) according to specific microscopic hypotheses (unfavorable, balanced or irreversible nucleation). We believe that our procedure could be applied to several related models (for instance, the Lifshitz-Slyozov-Wagner equation, previously mentioned, or the prion equation [12]) and should help to build reduced structured population models while taking into account of their intrinsic multi-scale nature (see [34,35] for applications).

Appendix: From the original to the dimensionless BD system. The original BD model gives the evolution of $(c_i)_{i\geq 1}$ by

$$\frac{d}{dt}c_1 = -J_1 - \sum_{i=1}^{\infty} J_i\,,\ t\geq 0\,,$$
$$\frac{d}{dt}c_i = J_{i-1} - J_i\,, \qquad t\geq 0\,,\ i\geq 2\,,$$

where J_i is the flux between clusters of size i and $i+1$, given by

$$J_i = a_i c_1 c_i - b_{i+1} c_{i+1}\,, \quad i\geq 1\,.$$

Here, coefficients a_i and b_{i+1} denote respectively the rate of aggregation and the rate of fragmentation. Observe that such model (at least formally) preserves the total number of particles (no source nor sink), that is

$$\sum_{i=1}^{\infty} i c_i(t) = \sum_{i=1}^{\infty} i c_i(0) =: m\,, \quad t\geq 0\,.$$

The classical approach to operate a scaling is to write the equations in a dimensionless form. We follow [9] and introduce the following characteristic values:

$\overline{T}$: characteristic time,
$\overline{C}_1$: characteristic value for the free particle concentration c_1 ,
$\overline{C}$: characteristic value for the cluster concentration c_i, for $i\geq 2$,
$\overline{A}_1$: characteristic value for the first aggregation coefficient a_1,
$\overline{B}_2$: characteristic value for the first fragmentation coefficient b_2,
$\overline{A}$: characteristic value for the aggregation coefficients a_i, $i\geq 2$,
$\overline{B}$: characteristic value for the fragmentation coefficients b_i, $i\geq 3$,
$\overline{M}_c$: characteristic value for the total mass m.

Thus, the dimensionless quantities are

$$\tilde{t} = t/\overline{T}\,, \quad \tilde{m} = m/\overline{M}_c\,, \quad \tilde{u}(\tilde{t}) = c_1(\tilde{t}\overline{T})/\overline{C}_1\,, \quad \tilde{c}_i(\tilde{t}) = c_i(\tilde{t}\overline{T})/\overline{C}\,,$$

and for all $i\geq 2$,

$$\tilde{a}_i = a_i/\overline{A}\,, \quad \tilde{b}_{i+1} = b_{i+1}/\overline{B}\,,$$

and the particular scaling at the boundary (we use different letters to emphasize this point):

$$\tilde{\alpha} := a_1/\overline{A}_1\,, \quad \tilde{\beta} := b_2/\overline{B}_2\,.$$

Then, the quantities $\tilde{u}(\tilde{t})$, $\tilde{c}_i(\tilde{t})$ satisfy the equation

$$\begin{aligned}
&\frac{d}{d\tilde{t}}\tilde{u} = \frac{\overline{C}}{\overline{C}_1}\Big[-\overline{AC}_1\overline{T}\Big(2\frac{\overline{A}_1\overline{C}_1}{\overline{AC}}\tilde{\alpha}\tilde{u}^2 + \sum_{i\geq 2}\tilde{a}_i\tilde{u}\tilde{c}_i\Big) + \overline{BT}\Big(2\frac{\overline{B}_2}{\overline{B}}\tilde{\beta}\tilde{c}_2 - \sum_{i\geq 3}\tilde{b}_i\tilde{c}_i\Big)\Big]\,,\\
&\frac{d}{d\tilde{t}}\tilde{c}_2 = \overline{AC}_1\overline{T}(\frac{\overline{A}_1\overline{C}_1}{\overline{AC}}\tilde{\alpha}\tilde{u}^2 - \tilde{a}_2\tilde{u}\tilde{c}_2) - \overline{BT}(\frac{\overline{B}_2}{\overline{B}}\tilde{\beta}\tilde{c}_2 - \tilde{b}_3\tilde{c}_3)\,,\\
&\frac{d}{d\tilde{t}}\tilde{c}_i = \overline{AC}_1\overline{T}(\tilde{a}_{i-1}\tilde{u}\tilde{c}_{i-1} - \tilde{a}_i\tilde{u}\tilde{c}_i) - \overline{BT}(\tilde{b}_i\tilde{c}_i - \tilde{b}_{i+1}\tilde{c}_{i+1})\,, \quad i\geq 3\,.
\end{aligned}$$

The mass conservation reads

$$\tilde{u}(\tilde{t}) + \frac{\overline{C}}{\overline{C}_1}\sum_{i\geq 2} i\tilde{c}_i(\tilde{t}) = \frac{\overline{M}_c}{\overline{C}_1}\tilde{m}\,.$$

We introduce the scaling parameter $\varepsilon > 0$ for the size of the clusters. Namely, a cluster of size i is now seen as a cluster of size roughly εi so that we can define the density (1.6). Then, the scaling obtained in Eq. (1.1) corresponds to the following choice of relations between the characteristic values

$$\overline{C}/\overline{C}_1 = \varepsilon^2\,, \quad \overline{AC}_1\overline{T} = \overline{BT} = \frac{1}{\varepsilon}\,, \quad \overline{M}_c/\overline{C}_1 = 1\,,$$

and, at the boundary,

$$\overline{A}_1 = \varepsilon^2\overline{A}\,,$$

and

$$\overline{B}_2 = \varepsilon^\eta\overline{B}\,,$$

with $\eta \geq 0$. The reader interested in a physical justification of this scaling can refer to the discussion in Section 7 and to [9].

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