Weierstraß-Institut
für Angewandte Analysis und Stochastik

im Forschungsverbund Berlin e.V.

Preprint ISSN 0946 – 8633

Mean field diffusion models for precipitation in crystalline GaAs including surface tension and bulk stresses

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submitted: 22nd December 2009

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No. 1475
Berlin 2009

2010 *Mathematics Subject Classification.* 74Q99, 35B27, 35Q74, 35R35, 74A15, 74F99, 74N20, 65P99.

2008 *Physics and Astronomy Classification Scheme.* 02.30.Jr,61.72.uj,64.70.D-,66.30.-h.

Key words and phrases. mean field approximation, homogenisation, semi-insulating GaAs, Ostwald ripening including mechanics.

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Abstract

Based on a thermodynamically consistent model for precipitation in gallium arsenide crystals including surface tension and bulk stresses by Dreyer and Duderstadt, we propose different mathematical models to describe the size evolution of liquid droplets in a crystalline solid. The first class of models treats the diffusion-controlled regime of interface motion, while the second class is concerned with the interface-controlled regime of interface motion. Our models take care of conservation of mass and substance. We consider homogenised models, where different length scales of the experimental situation have been exploited in order to simplify the equations. These homogenised models generalise the well-known Lifshitz-Slyozov-Wagner model for Ostwald ripening. Mean field models capture the main properties of our system and are well adapted for numerics and further analysis. Numerical evidence suggests in which case which one of the two regimes might be appropriate to the experimental situation.

Industrial problem and modeling

Semi-insulating gallium arsenide has a broad range of applications in micro- and opto-electronic devices. The industrial production process of semi-insulating gallium arsenide (GaAs) as done by Freiberger Compound Materials requires at the end some additional final heat treatment at high temperatures ($\sim$ 1000 K – 1200 K) in order to improve the quality of the semi-insulator. During this treatment undesirable liquid droplets precipitate in the solid phase due to supersaturation. The precipitation process is accompanied by surface tension and mechanical bulk stresses due to misfits. Droplets negatively influence mechanical and semi-insulating properties of the crystal. Their elimination, if possible, is a crucial point for the production of semi-insulators.

One of the challenges is the necessity to guarantee a mean mole fraction of As in the wafer of $X_0 = 0.500082$, which is specified to an accuracy of $\mathcal{O}(10^{-6})$, in order to have the desired semi-insulating behaviour. Since experiments have to be carried out at high temperatures and high pressure, mathematical modelling is important in this situation in order to understand well the evolution of a large number of precipitates. The goal of a mathematical model that describes the nucleation and evolution of the precipitates is to look for regimes, where for large times either only a few relatively big droplets survive or where a homogeneous spatial distribution of relatively small droplets results.

A GaAs crystal has a fcc-lattice structure with three sublattices. We work with the

so-called reduced Freiberg model [8], [17], [6] in which Ga atoms only appear on one sublattice, while As atoms and Va (Vacancies) can occupy lattice sites in all three sublattices. In the reduced Freiberg model the doping by B, Si and O atoms and the free charges e, h are neglected.
For the modelling of phase transitions various types of models are suggested. Sharp-interface models e.g. as the Mullins-Sekerka model [13] and phase-field models e.g. the Cahn-Hilliard equation [3] capture the spatial structure of a phase transition, while Lifshitz-Slyozov-Wagner models (LSW) [12], [18] and Becker-Döring models [2], [9] do not. The first three of these models are diffusion models which consider a continuum, while the Becker-Döring model is a discrete model, which considers the droplets as clusters of a certain number of atoms, which shrink or grow by gaining or loosing a single atom.
Phase-transitions in solids are in general strongly influenced by surface tension and bulk stresses, which is not reflected by the above-mentioned classical models. Furthermore in the situation of a phase transition within a solid crystal, the microstructure of the crystal cannot be neglected. For crystalline GaAs a thermodynamically consistent sharp-interface model for precipitation of liquid droplets including surface tension, bulk stresses and the crystal microstructure has been derived by Dreyer and Duderstadt [6]. They assume constant temperature T and constant outer pressure p_0. They proposed a thermodynamically consistent Becker-Döring model (BD) in the same situation, too [5]. In particular they derive a BD-model with a thermodynamically consistent choice of the condensation and evaporation rates, which is different to the Becker-Döring models usually considered by mathematicians [1]. For rigorous mathematical results for the Dreyer-Duderstadt-Becker-Döring model we refer to Herrmann *et al.* [10]. However, comparison with experimental results, shows that the time scales of the Becker-Döring regime are too large in case of crystalline GaAs.
In the diffusion model of Dreyer and Duderstadt there a two possible regimes, a diffusion-controlled regime of interface motion (DC) vs. an interace-controlled regime (IC). In the DC-regime we can have the situation of a constant diffusion constant D or of a constant bulk mobility B^D. Since from experiments it is a priori not clear which of the regimes might be appropriate, we consider the regimes separately in our study.
The main assumptions in the diffusion models are, that we can deal in good approximation with $\mathcal{N}$ spherical liquid droplets with fixed droplet centers X_i, $1 \leq i \leq \mathcal{N}$. We neglect nucleation of new droplets. We concentrate on arsenic-rich liquid droplets in a GaAs crystal. The wafer is considered as a body, represented by an open bounded domain Ω in three dimensions, which is time-dependent. Droplets can shrink or grow with time. The free boundary of the phase interface of a droplet i is parametrised by its radius r_i. There is a certain minimal radius, below which a liquid droplet does not behave like a droplet any more. We model this by taking the droplet out and incorporating the As and Ga atoms of the droplet into the solid. For the modelling of this subtle point, see [11], Subsect. 2.6.7. The liquid droplets are describes as spherical balls $B_{r_i}(X_i)$ with radius r_i and center X_i. The solid phase is represeted by a simply connected open domain $\Omega_S := \Omega \setminus \cup_i \overline{B_{r_i}(X_i)}$. For our

notation of the geometric quantities, see also Fig. 1. In this study we ignore misfits of the crystal due to dislocations in the crystal structure and the corresponding mechanical eigenstresses.

If one neglects surface tension and bulk stresses the system is well-understood,

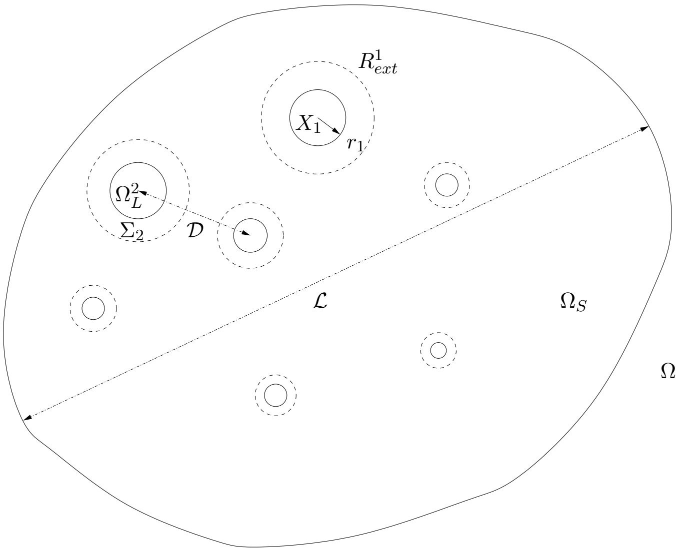

Figure 1: Many droplet problem with arbitrary convex shape. Dashed radii R^i_{ext} model the local influence of a droplet in a monopole approximation in a spherical shell Σ_i.

that can be realized by a solid cube of GaAs crystal in a liquid GaAs bath where the external pressure acts by means of a gas of As_2 and As_4 on the system (see [6], Fig. 2) such that the crystal is under hydrostatic pressure in 3-phase equilibrium. This is called the reference standard system and we consider actual values as deviations from the values of 3-phase equilibrium, which we indicate by an index R e.g. the vapour pressure under which three phase equilibrium may be established is the vapour pressure $p^R(T)$, while the real presure is p. We work in approximation of small displacement gradients in this study.

The differential equations derived by Dreyer and Duderstadt have been modified by Kimmerle ([11], Sect. 2) in a way, which is appropriate for mathematical analysis. The unknowns are the chemical potential μ, the mechanical displacement field u in Eulerian coordianates, the radii of the droplets r_i, and the free outer boundary $\partial\Omega$. By comparison of this modified differential equations with the Mullins-Sekerka model, we see that the inclusion of mechanical stresses and the microstructure yields in the solid crystal a quasi-linear parabolic diffusion equation with a drift term, which is coupled to a mechanical elliptic linear system by the barycentric velocity $v = \partial_t u$. In the evolution equation for the free boundary, the so-called Stefan condition, the mechanical phenomena enters, too. Contrary to classical models the outer boundary of solid crystal is a free boundary too, which evolves with the barycentric velocity v. The classical Gibbs-Thomsn law, which states that the chemical potential on the phase interfaces is given by surface tension σ times mean curvature, has to be replaced by a general strictly monotone decreasing, nonnegative and smooth function $\mu_I(r_i)$, which reflects the appearence of non-zero deviatoric stresses at the interfaces. In the liquid droplet i the chemical potential μ_L and the mechanical displacement in the liquid u_L are determined as a formula dependent on r_i and on the boundary displacement $u|_{I_i}$ on the interface I_i.

Due to different scales for typical distances between droplets and typical radii of liquid droplets (see Fig. 1) we expect that the chemical potential μ behaves far away from droplets approximately like an only time-dependent mean-field $\overline{\mu}$, while in the neighbourhood of a droplet with radius r_i then μ is dominated by $(\overline{\mu}(t) - \mu_I(r_i))r_i^{-1}$. The vector field of the displacement gradient ∇u is expected to behave like $(p^R - p_0)/(3k_S) = const$, where $k_S(T)$ is the bulk modulus of crystalline GaAs. Close to droplets we find $\nabla u \approx a + b(r_i)$ with $b(r)(4G_S + 3k_L) = 3(k_S - k_L)a - 2\sigma/r + \delta + h_L^*(r) - 3k_S(4G_S + 3k_L)/(4G_S + 3k_S)h^*(r)$. $\delta(T)$ is the misfit parameter and h^*, h_L^* model the contribution due to inelastic deformations from change of concentrations in the solid and liquid, resp. $G_S(T)$ is the shear modulus in the solid and $k_L(T)$ is the bulk modulus in the liquid. Furthermore the lattice occupancy Y^R of As atoms on the interstitial lattice in the reference system is small, so the material-derivative of the chemical potential and the coupling to the velocity enter in higher order in the diffusion equation.

We want to exploit the different scales in order to simplify our generalised Mullins-Sekerka model. Let $\mathcal{D}$ the typical distance of a droplet to the next neighbour droplet or to the ouer boundary $\partial\Omega$ and $\mathcal{L}$ the diameter of a homogeneous precipitation domain Ω, that is much larger than $\mathcal{D}$. We introduce the scaling parameter $\varepsilon = \mathcal{D}/\mathcal{L}$. This yields directly that the number of precipitates scales with ε^{-3}. $\varepsilon_0 = 10^{-1}$ is the value of the scaling parameter, which fits to the experiment. We give an overview of most of the used unscaled quantities and their scaling behaviour in Table 1. Mathematically the scaling behaviour corresponds to a dilute scaling regime i.e. radii are scaled with ε^4. Numerics yield the guess, that the dilute regime might fit to the experimental situation. Similarly as the LSW-model is derived as a homogenization limit of the Mullins-Sekerka model in the regime of small volume fraction, see [15], [16], we exploit the different length scales and derive from our thermodynamically consistent generalization of the Mullins-Sekerka model, a generalized LSW-model for precipitation in crystalline GaAs including mechanical stresses and the microstructure.

Mathematical results for both genealized models and the derivation of them are presented in [11]. For a model in the diffusion-controlled regime we prove the limit of small volume fraction in the critical scaling by homogenisation techniques under plausible assumptions in [11], Ch. 5.

Quantity	**Experimental value**	**Scaling**	**Scaled value**
Scaling parameter	$\varepsilon^0 = 10^{-1}$	ε	ε^0
Diameter of domain $\mathcal{L}$	10^{-5}m	$\mathcal{L}$	1
Typ. droplet dist. $\mathcal{D}$	10^{-6}m	$\varepsilon^1\mathcal{L}$	1
Typ. initial radii $\mathcal{R}$	10^{-9}m	$\mathbf{L} = \varepsilon^4\mathcal{L}$	1
Typical times (DC) τ^D	10^{-6}s	$\mathbf{T} = \tau^D$	1
Typical times (IC) τ^I	10^{-12}s	$\mathbf{T} = \tau^I$	1
Typical masses $\mathcal{M}$	M_{As} $= 7.492 \cdot 10^{-2}$kg mol^{-1}	$\mathbf{M}/\text{mol} = \varepsilon^4 M_{As}$	1
Initial # of droplets $\mathcal{N}^0$	$4.2 \cdot 10^3$	$(\frac{\mathcal{L}}{\mathcal{D}})^3 = \varepsilon^{-3}$	$4.2 \cdot 10^3$
Initial # of atoms N_0	$1.01 \cdot 10^{-10}$mol $\approx (3 - Y_V^R) n_G^R \mathcal{L}^3$	$\mathbf{N} = \varepsilon^{12}$ 1mol	$1.01 \cdot 10^{-10}$
Ref. density of lattice sites n_G^R	$3.7 \cdot 10^4$mol m^{-3}	ε^0 1mol m^{-3}	$3.7 \cdot 10^4$
Ref. density of atoms in the lq. n_L^R	$3.7 \cdot 10^4$mol m^{-3}	ε^0 1mol m^{-3}	$7.0 \cdot 10^4$
Init. density of droplets $N_0^R = \frac{N_0}{\mathcal{N}^0}$	$2.4 \cdot 10^{-14}$mol	$\mathbf{N} = \varepsilon^{15}$ 1mol	$2.4 \cdot 10^{-14}$
Ref. concentr. of As$_\gamma$ Y^R	10^{-4}	ε^9	10^{-4}
Mobility (DC) B^D	$3.7 \cdot 10^{-14}\,\frac{\text{mol}}{\text{m s}}$	$\frac{\mathbf{N}}{\mathbf{LT}} \propto \varepsilon^8$	$3.7 \cdot 10^{-23}$
Mobility (IC) B^I	$9.8 \cdot 10^6\,\frac{\text{mol}}{\text{m}^2\,\text{s}}$	$\frac{\mathbf{N}}{\mathbf{L}^2\mathbf{T}} \propto \varepsilon^4$	$9.8 \cdot 10^{-24}$
Diffusion coefficient (DC) D	$10^{-12}\,\frac{\text{m}^2}{\text{s}}$	$\frac{\mathbf{L}^2}{\mathbf{T}} \propto \varepsilon^8$	1
Typ. stresses e.g. $k_S k_L, G_S$	$10^7\,\frac{\text{N}}{\text{m}^2}$	$\frac{\mathbf{M}}{\mathbf{LT}^2} \propto \varepsilon^0$	10^7
Surface tension σ	$7.5 \cdot 10^{-2}\,\frac{\text{N}}{\text{m}}$	$\frac{\mathbf{M}}{\mathbf{T}^2} \propto \varepsilon^4$	0.075
Typ chem. pot. u	$RT = 9.14 \cdot 10^3$J mol^{-1}	$\frac{\mathbf{ML}^2}{\mathbf{NT}^2} \propto 1$	$9.14 \cdot 10^3$

Table 1: Overview of typical quantities for the experiment, the scaling and the scaled values for DC or IC.

Mean field models

From this generalized LSW-model, consisting of a kinetic equation and an integro-differential equation for the mean field $\overline{\mu}$, we get a mean field model by considering an initial measure, which consists of a finite number $\mathcal{N}^0$ of initially existing droplets with initial radii r_i^0. In order to guarantee conservation of mass and substance we consider in the mean field models $|\Omega_S(t)|$ instead of $|\Omega(0)|$ as a formal next-order corrector and evaluate ε for the corresponding scale of a typical experimental system. We consider as well the regime DC as well the regime IC. Our mean field problems are the following coupled nonlinear ODE systems for μ and r_i

$$\dot{r}_i(t) = G(r_i(t), \overline{\mu}(t)) \quad \forall i \in \{1, ..., \mathcal{N}(t)\}, \quad \dot{\overline{\mu}}(t) = H(\{r_i(t)\}_{1 \le i \le \mathcal{N}}, \overline{\mu}(t)), \tag{1}$$

where in the regime DC in case of constant diffusion coefficient

$$G := D\frac{\overline{\mu}(t) - \mu_I(r_i)}{r_i\overline{\mathbb{X}}(r_i)}, \quad H := -4\pi D\frac{1}{\mathcal{N}^0}\frac{\sum_{i\in N} r_i(\overline{\mu}(t) - \mu_I(r_i))}{Y^R\overline{\mathcal{X}}|\Omega_S(\{r_i(t)\},\overline{\mu})|}, \tag{2}$$

while in the regime IC

$$\begin{aligned} G &:= \frac{B^I}{RTn_G^R}\frac{\overline{\mu}(t) - \mu_L(\overline{\mu}, r_i)}{\overline{\mathbb{Z}}(\overline{\mu}, r_i)}, \\ H &:= -4\pi\frac{B^I}{RTn_G^R}\frac{1}{\mathcal{N}^0}\frac{\sum_{i\in N} r_i^2(\overline{\mu}(t) - \mu_L(\overline{\mu}, r_i))\mathbb{Y}(r_i,\overline{\mu})}{Y^R\overline{\mathcal{X}}|\Omega_S(\{r_i(t)\},\overline{\mu})|}, \end{aligned} \tag{3}$$

together with initial conditions on r_i and $\overline{\mu}$. $\mathbb{X}$ or $\mathbb{Z}$ model the contribution of the barycentric velocity in the solid or liquid at the phase boundary, which are eliminated in the Stefan condition for regime DC or IC. By these two functions the mechanical bulk stresses at the interface enter into the equations. $\mathcal{X}$ represents the derivative of the concentration of the As concentration in the solid w.r.t. μ and gives the influence of the microstructure in the equations. The functions $\mathbb{X}$, $\mathbb{Z}$, $\mathbb{Y}$ and $\mathcal{X}$ are of order 1. The outer boundary $\partial\Omega$ and the size of the solid domain $|\Omega_S|$ are determined by conservation of mass and substance. The change of the outer boundary is small compared to the change of radii.

Furthermore it can be checked formally, that the available free energy corresponding to the mean field model is non-increasing with time, see [11]. In this sense the mean field model is thermodynamically consistent.

Mean field models as (1) – (3) are more suitable for simulations and stability analysis than the original coupled PDEs and ODEs of a generalized Mullins-Sekerka model. We recall that the boundary condition on the interfaces of the droplets, $\mu_I(r_i)$, is a strictly monotone decreasing and nonnegative smooth function of r_i. This suggests to replace equivalently the mean field $\overline{\mu}$ by a mean field radius $\overline{r} = \mu_I^{-1}(\overline{\mu})$. This is similar as in the situation of the classical LSW-model where $\mu_I(r_i) = 1/r_i$ and $\overline{r} = 1/\overline{\mu}$. $\overline{r}$ turns again out to be the critical radius. That means, that droplets with radii below $\overline{r}$ shrink, while radii beyond grow.

Mean field models now consist of $\mathcal{N}^0$ nonlinear ODEs for the evolution of the radii r_i, $1 \le i \le \mathcal{N}^0$, and one ODE for the mean field radius $\overline{r}$, closed with initial conditions r_i^0, $1 \le i \le \mathcal{N}^0$, and $\overline{r}^0$. W.l.o.g. $r_1 > r_2 > ... > r_{\mathcal{N}^0}$. The dissolution of droplets with time is modelled as follows. Let $\mathcal{N}(t)$ be the number of existing droplets at time t. If $r_{\mathcal{N}} \le r_{min}$ for a given value r_{min}, which is motivated from physics, at time $\tau_{\mathcal{N}}$, then $\mathcal{N} - 1$ droplets remain and the mean field model has after $\tau_{\mathcal{N}}$ hence $\mathcal{N} - 1$ equations for the radii and one for the mean field, with new initial data, which is given from the values at time $\tau_{\mathcal{N}}$.

Furthermore the initial condition $\overline{r}(0) = \overline{r}^0$ can be replaced by prescribing $N_0 = \mathcal{N}^0 N_0^R$, where N_0^R is the typical number of atoms in a box with side length $\mathcal{D}_0 \approx 10^{-6}m$ around the droplet center, where the atoms in the droplet are also included in N_0^R.

The change of radii depends on the sign of $r_i - \overline{r}$. Radii grow, if this difference is positive, radii shrink, if $r_i < \overline{r}$ or remain stationary if the difference is zero.

Stationary solutions of mean field models

We determine possible stationary solutions i.e. where all time-derivatives in the ODE system vanish, and in the case of $1 \leq \mathcal{N}(t) \leq \mathcal{N}^0$ we see that this is only possible iff $r_i = \overline{r}$, for all droplets $1 \leq i \leq \mathcal{N}$. In case of $\mathcal{N} = 0$ the mean chemical potential $\overline{\mu}$ remains clearly stationary.
These stationary solutions for the radii (or denoted as "equilibria" in [11]) can be determined for any number of remaining droplets $\mathcal{N}$. For typical experimental situations two different stationary radii are possible, $r^{\mathcal{N}}_{crit}$, which turn out to be instable, or $r^{\mathcal{N}}_{ms}$, which turn out to be instable for $\mathcal{N} > 1$, but is asymptotically stable if $\mathcal{N} = 1$. The system is stable in the sense of Lyapunov, see e.g. [19], for $\mathcal{N} = 0$. The stability of the stationary solutions of the ODE system is here examined by means of linearisation around stationary solutions and explicit computation of the corresponding eigenvalues of the linearised system (Theorem of Poincaré-Lyapunov), see [11], Sect. 6.3.
Analoguesly the homogenised model can be put into the original formula for the availabile free energy and we can analyse this availabile free energy $\mathcal{A}$ in case of the mean field model. The stationary solutions correspond to extrema of the formally homogenised availabile free energy. $r^{\mathcal{N}}_{crit}$ corresponds to maxima of the availabile free energy of the mean field model, while $r^{\mathcal{N}}_{ms}$ corresponds to a saddle point i.e. a metastable state for $\mathcal{N} > 1$ and to a minima, which is stable, if $\mathcal{N} = 1$ or 0. In case of two droplets this is illustrated in Fig. 2.

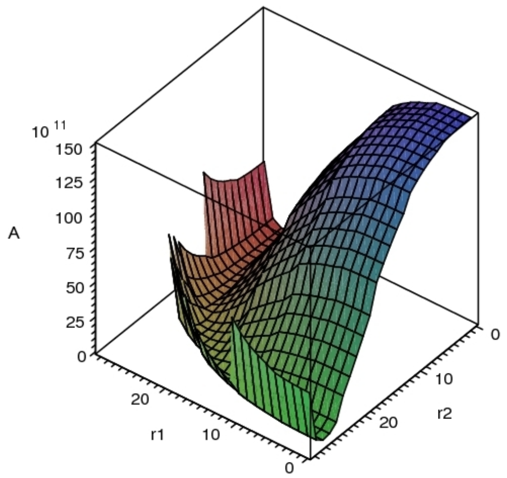

Figure 2: Availabile free energy [in 10^{-27} N m] versus radii [in 10^{-9} m] for the mean field model of a system with $\mathcal{N}^0 = 2$ and here $N^R_0 = 2.39 \cdot 10^{-14}$mol, $p_0 = 10^5\,\mathrm{N\,m^{-2}}$. The equilibrium $(r_1 = r^2_{crit}, r_2 = r^2_{crit})$ is unstable, $(r_1 = r^1_{crit}, r_2 = 0)$, $(r_1 = 0, r_2 = r^1_{crit})$ and $(r_1 = r^2_{stab}, r_2 = r^2_{stab})$ are metastable saddle points and $(r_1 = r^1_{stab}, r_2 = 0)$, $(r_1 = 0, r_2 = r^2_{stab})$ are stable equilibria.

Numerical solutions of the mean field models

We see in simulations the following typical behaviour, which is expected due to the analysis of stationary solutions. The mean field radius runs with time close to a quasi-stationary equilibrium, which is unstable (i.e. a "saddle") (see [11], Fig. 6.13). The smallest droplet vanishes, then the next saddle is approached, and so on, unless only one droplet remains, and the mean field approaches the radius r_1 of the remaining droplet and this situation is stable.
For typical situations the change of the mean field is faster than the change of radii and the mean field radius approaches a meta-stable value $\overline{r} \approx \frac{1}{\mathcal{N}^0} \sum_{1 \leq i \leq \mathcal{N}} r_i$.
We remark, that our mean field model can be interpreted in the sense, that the

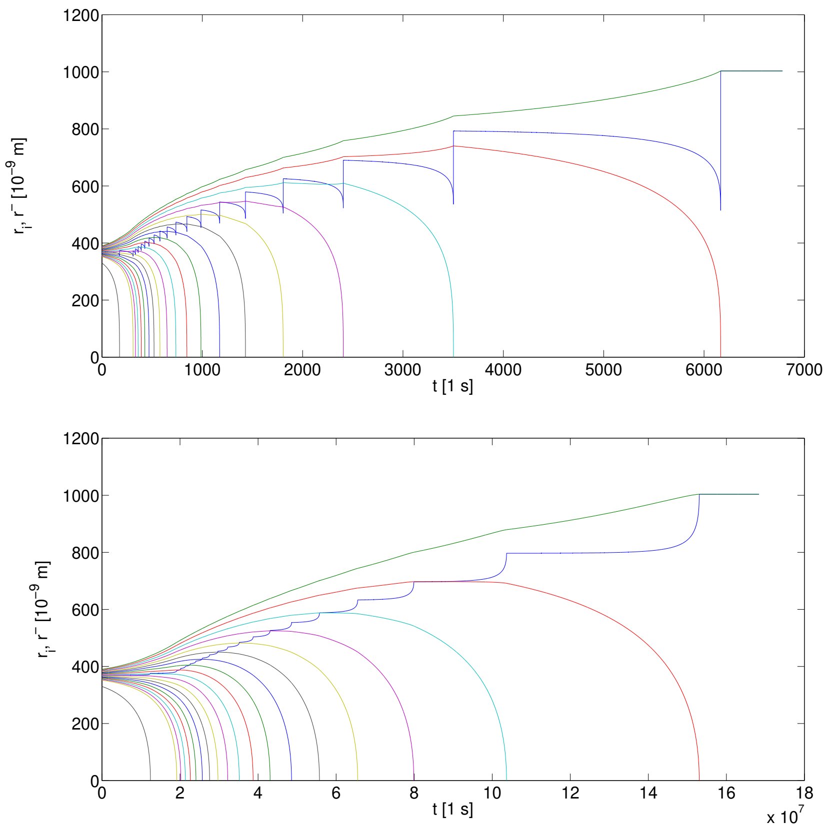

Figure 3: Simulations of mean field models: on the l.h.s. regime DC with $D = 10^{-12}\mathrm{m}^2\mathrm{s}^{-1}$, on the r.h.s. regime IC with $B^I = 10^{-5}\mathrm{mol\ m}^{-2}\mathrm{s}^{-1}$. In both cases 1 droplet (darkgreen) remains and mean field radius (darkblue) is plotted for all times, all other droplets dissolve. Both simulations with identical initial data: 20 radii uniformly distributed between 320-400 nm, $N_0^R = 3.8 \cdot 10^{-14}$ mol, $T = 1100$K and the data from Table 1.

"information" about the position of the initial radii gets lost, when the dynamical system runs for the first time close to a saddle.

Except for special initial data e.g. if all initial droplets are below the mean field radius $\overline{r}$ and sufficiently small enough so that they vanish before the mean field runs into a quasi-stationary equilibrium,we end up with one droplet.
We summarise. If we start with a fixed number of $\mathcal{N}^0$ droplets, we could theoretically end up with no droplet, 1 droplet or $2 \leq \mathcal{N} \leq \mathcal{N}^0$ droplets. More than one droplet is only possible for very pathological cases, where the initial data is a critical point or saddle or leads perfectly into a saddle and that cannot be realised practically. Simulations show, that for realistic initial data it can be expected, that 1 droplet remains as $t \to \infty$.

We consider the life time τ_i of droplet i. The time $\tau_{\mathcal{N}}$ until the first droplet disappears which can be read off from plots, can be compared with times from experiments. This offers a possibility to decide which model might be appropriate for the experimental situation.
Numerical evidence suggests in which case which one of the two regimes might be appropriate to the experimental situation: The IC regime with $B^I = 10^{-5}$mol m^{-2}s^{-1}, which is an artificially large chosen value, already yields times τ_1 and $\tau_{\mathcal{N}^0}$, which seem to be too large. The DC regime, where $B = 3.7 \cdot 10^{-8}$mol m^{-1}s^{-1} is kept constant and not D, yields too short times. The DC regime with $D = const = 10^{-12}$m^{2}s^{-1} might be the most appropriate. Furthermore note, that the mean field radius exhibits a qualitatively different behaviour for regimes DC and IC before and after a droplet dissolves.
The simulations still depend sensitively on values for the diffusion constant or mobilities and surface tension.
As a further result, which has not been included in the PhD thesis of Kimmerle [11], the dependence of the droplet evolution of parameters can be characterised approximately as follows. $\tau_1 \sim \frac{1}{\sqrt{\sigma D}}$ for DC and given constant diffusion coefficient D, while $\tau_1 \sim \frac{1}{\sigma B^I}$ for regime IC. Simulations suggest, that the more droplets we consider, the smaller the life times $\tau_{\mathcal{N}}$. The shape of the initial radii data and N_0^R have influence on how fast a quasi-stationary equilibrium is reached and where a quasi-stationary radius (the radius corresponding to a saddle of the availabile free energy) is exactly. The more concentrated the initial data is around a point, the larger we expect τ_1 and $\tau_{\mathcal{N}}$.
The effect of the external pressure p_0 is not significant for simulations as can be expected from the analysis. The most significant effect of the temperature seems to be the dependence of $Y^R(T)$: the smaller Y^R, the faster the change of mean field radius compared to change of droplet radii.
Numerical results suggest the existence of self-similar solutions for our mean field models, see Fig. 4. For classical LSW models the existence of self-similar solutions is possible and is well-understood, depending on the initial data. This is a priori not clear for our generalized LSW models, since we have a further dependence on the radii in the functions $\mathbb{X}$ for DC or $\mathbb{Y}$ and $\mathbb{Z}$ for IC.

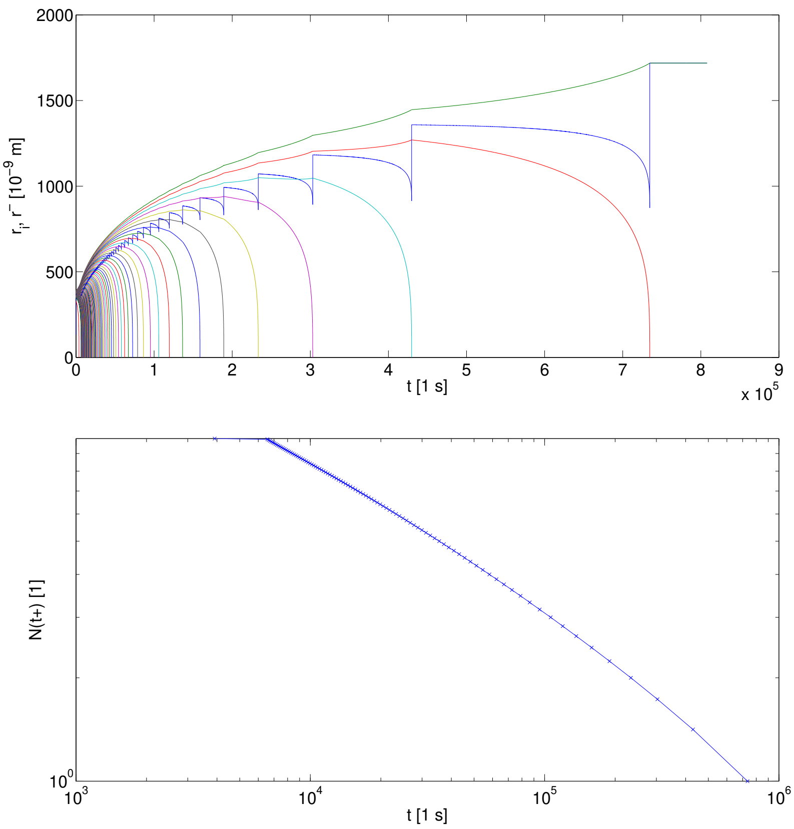

Figure 4: Regime DC, 100 droplets, Simulation (r.h.s.) and $\mathcal{N}$ vs. time t on a log-log-scale (l.h.s.)

Conclusion

Finally we compare our model with other models, like the Becker-Döring model [5], [10] or models for grow and shrinkage of rubber balloons or storage processes in rechargeable lithium batteries (see e.g. [7]).
In case of BD-model the time scales for dissolving droplets are too large compared to experiments. The availabile free energy, which is used as starting point in our model has no mixing entropy term, contrary to the availabile free energy for the Becker-Döring system.
We point out, that in our case μ_I is strictly monotone, and the mean field is uniquely determined by an ODE (our equivalently by a monotone nonlinear equation for $\overline{\mu}$). In the models for rubber balloons and lithium batteries hysteresis can occur since $\overline{\mu}$ has two distinct values (depending on the scaling a bifurcation occurs or not), which are selected depending on the history of the system and/or by an external control. Another question is, how a homogenised model might look like for these more complicated models exhibiting hysteresis and non-monotonic behaviour.
A further goal for the near future is the better understanding and simulation of

the model including the full kinetic equation, which allows to understand the case of many droplets, since simulations of mean field models seem to be feasible only for a relatively small number of droplets (up to $100 - 500$ droplets). A numerical approach for the classical LSW model has been first given by Carillo and Goudon [4]. Our aim is to follow an ansatz of Peschka et al. [14], which consider a thin film equation of LSW type. As in this case we have to carefully dissolve the maximal radius in our numerics.

Acknowledgement

The authors would like to thank Stefan Eichler and Manfred Jurisch for many fruitful discussions and for their valuable comments and remarks. S.-J. Kimmerle acknowledges gratefully support through the DFG research center MATHEON.

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