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Convergence, parameter stability and simulation

Nicklas Jävergård

Faculty of Health, Science and Technology

Mathematics

LICENTIATE THESIS | Karlstad University Studies | 2026:21

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urn:nbn:se:kau:diva-109263

ISSN 1403-8099

ISBN 978-91-7867-690-3 (print)

ISBN 978-91-7867-691-0 (pdf)

<https://doi.org/10.59217/aqna3840>

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Distribution:

Karlstad University

Faculty of Health, Science and Technology

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SE-651 88 Karlstad, Sweden

+46 54 700 10 00

Print: Universitetstryckeriet, Karlstad 2026

WWW.KAU.SE

Finite volume schemes for a conservative hydrodynamic limit of the Kac-Blume-Capel model: Convergence, parameter stability and simulation

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Abstract

Morphology formation in thin films produced from a ternary solution is crucial for the performance of organic solar cells. Both the separation of excitons into free charges as well as the charge transport that follows depend on the shape and connectivity of the distinct polymer regions (the morphology). In this thesis, we study morphology formation from two different perspectives: A lattice-based Blume-Capel model with Kawasaki dynamics, and then a continuum system of coupled parabolic equations with nonlinear and nonlocal drift. The objective of this licentiate thesis is to represent morphology formation in three space dimensions using these two models. We relate our work to previous two-dimensional results for different parameter regimes. At the technical level, we construct a semi-discrete finite volume scheme to approximate the weak solution of our continuum model and implement it in `Julia`. We prove a convergence result of our semi-discrete scheme as well as a stability result of the weak solution with respect to temperature variations - a key parameter in the model. Looking at both the lattice model and the continuum parabolic system, we quantify and compare growth rates of the formed domains. Finally, we perform numerical experiments confirming convergence of our scheme and the effect of parameters on the obtained solution. These results provide a solid foundation for future extensions, including the evaporation of a mixture component.

Keywords: morphology formation, ternary mixture, domain growth, nonlinear parabolic system, nonlocal drift, weak solution, finite volume approximation, Blume-Capel model, Kawasaki dynamics

MSC (2020): 82C20, 35K55, 35R09, 35Q70, 65M08, 65M12, 65M22

Sammanfattning

Morfologibildning i tunna filmer framställda från en trenär lösning är avgörande för prestandan hos organiska solceller. Både separationen av excitoner till fria laddningar samt den efterföljande laddningstransporten beror på formen och konnektiviteten hos de distinkta polymerregionerna (morfologin).

I denna avhandling studerar vi morfologibildningen ur två olika perspektiv: en gitterbaserad Blume-Capel modell med Kawasaki-dynamik, och därefter ett kontinuumssystem av kopplade paraboliska ekvationer med icke-linjär och icke-lokal drift. Målet med denna licentiatavhandling är att representera morfologibildning i tre rumsdimensioner med hjälp av dessa två modeller. Vi relaterar vårt arbete till tidigare tvådimensionella resultat för olika parameterregioner.

På den tekniska nivån konstruerar vi ett semidiskret finita volym-schema för att approximera den svaga lösningen av vår kontinuummodell och implementerar den i Julia. Vi bevisar ett konvergens resultat för vårt semidiskreta schema samt ett stabilitets resultat för den svaga lösningen med avseende på temperaturvariationer – en nyckelparameter i modellen. Genom att studera både gittermodellen och det paraboliska kontinuumssystemet kvantifierar och jämför vi tillväxthastigheter för de bildade domänerna. Slutligen utför vi numeriska experiment som bekräftar konvergens hos vårt schema samt parameterarnas inverkan på den erhållna lösningen. Dessa resultat ger en solid grund för framtida utvidgningar, inklusive avdunstning av en blandningskomponent.

Acknowledgements

Throughout my studies I have met many people that have changed the way I think about almost everything. All were important in different ways and, without wishing to overlook anyone, I would like to mention a few in particular.

Adrian Muntean, you are the perfect person to be my supervisor. Your enthusiasm, knowledge and support have been invaluable for my development and the progress of my work. It has been and still is a great pleasure to be working with you. I would also like to thank the rest of my supervisory team, Grigor Nika, Pei Huang and Andreas Theocharis.

Andrea Stela Muntean, it so happened that we met early on in my studies. I constantly interrupted your day to day with, what I can only assume to be, annoying questions. You always took your time to listen, ask and encourage. Thank you for your help, encouragement and friendship.

I was very fortunate during my studies, both under graduate and graduate, as an under grad I had a tight group of friends that worked hard together. I would like to thank you, Daniel, Olof, Andreas, and Henrik, for the effort we did together and you continued (and unfounded) belief that I can do anything.

When I began my graduate studies I was greeted by a vibrant research group with amazing individuals. Rainey, Michael, Vishnu and Surrendra, for all the time you spent answering this former physicists rather silly questions, I thank you. More importantly, I thank you for your friendship and for mercilessly beating me at chess over and over and ... and over again.

Emilio Cirillo, Sapienza University of Rome, I want to thank you for hosting me in Rome and all the subsequent discussion and collaboration that followed. I would also like to express my gratitude to you for your openness and willingness to share and discuss your ideas.

Hirofumi Notsu, Kanazawa University, thank you for taking the time during your short visit to listen to my ideas and clarifying what parts of them that made sense.

Grigory Panasenko, University of Lyon/University of Saint-Etienne, thank you for

visiting us and sharing your work on boundary layer solutions as well as taking the time to discuss my research problems. Your insight and help proved very valuable.

Sander Hille, Leiden University, thank you for sharing your knowledge and enthusiasm. I am sure that these discussions will prove fruitful.

The main part of my work has been to develop computational codes and schemes to solve nonlinear, nonlocal PDEs. This can be a frustrating process, but I got great advice and help from colleagues at different universities. For instance, Sorin Pop from the University of Hasselt and Omar Lakkis from the University of Sussex.

To conclude these acknowledgments, I would like to thank my family—my mother, my father and my sisters—for their unbounded support and understanding. Lastly, I express my deepest gratitude to my fiancée Katerina Chantziara, you not only make this work possible, you make the time outside of my work fantastic!

I would be remiss if I did not acknowledge the partial financial support from the Swedish Energy Agency through the project Solar Electricity Research Centre (SOLVE), grant number 52693-1. The computations were enabled by resources provided by the National Academic Infrastructure for Supercomputing in Sweden (NAISS), partially funded by the Swedish Research Council through grant agreement no. 2022-06725, via the projects NAISS 2025/22-1655, NAISS 2025/3-65, and NAISS 2025/22-1096.

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Chapter 1

Introduction

1.1 Our motivation

The strive for affordable, abundant energy with low environmental impact is a topic widely discussed and its importance can not be overstated. The question is how can the energy needs of the world be satiated whilst reducing the strain on our shared environment? Photovoltaic (PV) technology is one proposed answer to this question. The total capacity of installed PV systems is significant and it is projected to increase in the future [1]. Although crystalline solar cells are considered “clean” energy, they still have a higher impact in terms of green house gas emissions per kWh when considering the entire manufacturing process compared with, for instance, hydropower; see [15].

In the recent years, the efficiency of organic solar cells (OSCs) have increased with values as high as 18% was reported in 2020, see [11]. These higher efficiencies, improvements in stability and low green house gas emissions now makes a compelling case for OSCs in the future. The photoactive layer of the prevailing OSCs consists of a bulk-heterojunction manufactured using a ternary mixture of a donor and acceptor phase immersed in a solvent. As evaporation happens in the physical system, the two competing phases separate and produce morphologies. The resultant morphology plays a vital role in the operation of the finished device. It is interesting to note however, that no one knows which shapes would result in optimal performance. The interface between the two polymer species is where free charges are generated by separating excitons. At the same interface, free charges can recombine and thus the charges are lost. The number of free charges generated and destroyed depends, among other things, on the available surface area and local shape of the phases, which influence the device stability. Furthermore, the distance of free charges to either anode or cathode affects the performance of a finished device. The interaction of the ternary mixture is the main objective of this thesis. Specifically, we wish to capture its behavior by means of mathematical models and

numerical simulations, thereby providing access to explore the produced shapes and connectivity (the morphology).

We are interested in those mathematical models that can accurately describe, to the extent possible, phenomena occurring during phase separation and the evolution toward morphologies observed in experiments. Ideally, we want to investigate how parameters of the models influence the resultant morphology, characterize it and, if possible, have a computational way to classify different morphologies. Such a classification would permit one to simulate charge on already available morphologies. Moreover, such simulations would offer a straightforward way to segregate more efficient shapes for the generating and extraction of free charges.

1.2 Structure of the thesis

This thesis is organized in the following way. In what follows of Chapter 1 we give a short description of the mathematical background of this thesis. Concretely, we briefly present the Blume-Capel model and its hydrodynamic limit. We present the goal of the thesis and highlight some challenges that the goal entails. As a final part of Chapter 1, the author presents an experiential account of his work from a computational perspective. In Chapter 2, the key findings of paper I and II are discussed separately. This Chapter also contains a list of publications and the author's contributions to the papers presented in this thesis. A list of proposed future work is given in Chapter 3. The remaining parts of this thesis contains our two published papers on this topic.

1.3 Description of our models

During our investigation of morphology formation in ternary mixture we have made use of two conceptually distinct models. One is a stochastic lattice-based Blume-Capel model with Kawasaki dynamics. The other is a deterministic continuum model derived via a hydrodynamic limit of the Kac-Blume-Capel model. In this section we will briefly describe them both. Although it is possible to work with these models in one-, two-, or three- spatial dimensions, within this thesis we focus exclusively on the three dimensional setting.

1.3.1 3D Morphology formation via a spin lattice model

We consider our lattice to be three dimensional. We refer to its elements as sites, the Euclidean distance between any two sites is denoted by $|i - i'|$ for $(i, i') \in \mathbb{Z}^3 \times \mathbb{Z}^3$. Given $i \in \mathbb{Z}^3$, the site $i' \in \mathbb{Z}^3$ will be referred to as the nearest neighbor of site i if and only if $|i - i'| = 1$. Each pair of nearest neighbors is called a bond. Let Λ be the

cubic torus $\{1, \dots, L-1\}^3 \subset \mathbb{Z}^3$, we associate with each site $i \in \Lambda$ a spin given by $\sigma(i)$ taking values $\{-1, 0, 1\}$. The collection of all spins in Λ is called a state space and is denoted by $\mathcal{X} = \{-1, 0, 1\}^\Lambda$. The energy of a given configuration $\sigma \in \mathcal{X}$ is given by the Hamiltonian function, $H : \mathcal{X} \rightarrow \mathbb{R}$, via

$$H(\sigma) := J \sum_{\langle i, j \rangle} [\sigma(i) - \sigma(j)]^2, \quad (1.1)$$

In (1.1), we set $J > 0$, representing the nearest neighbor interaction energy, while the sum is taken over $3L^3$ bonds with periodic boundary conditions. The function $H(\cdot)$ arising in equation (1.1) is here referred to as the Hamiltonian of the Blume-Capel model [2, 3]. In our polymer interpretation, the ± 1 refer to two different polymer species and 0 refers to the background solvent. The Blume-Capel model can also be endowed with different dynamics, conservative (Kawasaki) or not conservative (Glauber). Since we are using this model to describe the phase separation of polymers, the natural choice of dynamics is the Kawasaki one as this locally conserves ‘mass’. With Kawasaki dynamics at hand, the Hamiltonian will produce an energetic cost of $4J$ for a direct interface between a minus and a plus while an interface between zero and plus, or a minus, costs J . On the other hand, an interface between two sites with the same value will have zero contribution. This makes the model a good choice for studying the morphology formation of two polymers which strongly repel each other in the presence of a background solvent acting as a shield between the two phases.

The simulation is initialized where each site has c_0 probability of being 0 and $c_{+1} = c_{-1} = (1 - c_0)/2$ are the probabilities of a site being occupied with either $+1$ or -1 , respectively. The stochastic model proposed is a Markov chain, $\sigma_t \in \mathcal{X}$, with transition probability defined through the following procedure. At each time $t = 0, 1, 2 \dots$, one bond in σ_t connecting two sites with different spins are chosen at random. Then, the two spins are swapped with probability one if the energy associated with the swap, Δ , is non-positive or with probability $e^{-\beta\Delta}$ otherwise. Where the parameter β is interpreted as the inverse temperature of the system. In this way the configuration σ_t is formed. After $3L^3$ bonds have been updated, we say that one iteration of the system has been made.

1.3.2 3D Morphology formation via a system of nonlinear differential equations with nonlocal drift

The model presented in section 1.3.1 is a stochastic lattice (discrete) model. In this section, we consider a deterministic continuum model posed in a cubic domain $\Omega \subseteq \mathbb{R}^3$, $\Omega \neq \emptyset$. Our particular choice is the hydrodynamic limit for the Blume-Capel model with Kawasaki dynamics as presented in [13] with the modified Hamiltonian

$H_\gamma : \mathcal{X} \rightarrow \mathbb{R}$, defined via

$$H_\gamma(\sigma) := \frac{1}{2} \sum_{i \neq j \in \Lambda} J_\gamma(i-j)[\sigma(i) - \sigma(j)]^2 - \lambda \sum_{i \in \Lambda} [\sigma(i)]^2 - h \sum_{i \in \Lambda} \sigma(i), \text{ for all } \sigma \in \mathcal{X}. \quad (1.2)$$

Here $\lambda, h \geq 0$. Looking at the structure of this Hamiltonian $H_\gamma(\cdot)$, we refer to $J_\gamma : \mathbb{R}^3 \rightarrow \mathbb{R}$ as the Kac potential function, which assumes a particular scaling

$$J_\gamma(r) = \gamma^3 J(\gamma r), \quad (1.3)$$

for all $r \in \mathbb{R}^3$. For the function J , we impose the following conditions: $J \in C^2(\mathbb{R}^3)$, $J(r) = J(-r)$, $\int J(r) dr = 1$ and $J(r) = 0$ for all $r \in \mathbb{R}^3$ such that $|r| > 1$. The parameter γ scales the range of interaction. With these choices for the Kac potential and Hamiltonian with $\lambda = h = 0$ one arrives at the following system of partial differential equations for the pair (m, ϕ)

$$\begin{cases} \partial_t m = \nabla \cdot [\nabla m - 2\beta(\phi - m^2)(\nabla J \star m)], & \text{in } (0, T) \times \Omega, \\ \partial_t \phi = \nabla \cdot [\nabla \phi - 2\beta m(1 - \phi)(\nabla J \star m)], & \text{in } (0, T) \times \Omega, \end{cases} \quad (1.4)$$

endowed with periodic boundary conditions at $\partial\Omega$ for both unknowns and with the initial data

$$m(t=0) = m_0, \quad \phi(t=0) = \phi_0, \text{ in } \bar{\Omega}. \quad (1.5)$$

The parameter β that appears in (1.4) represents the inverse temperature. In (1.4), x and t are the space and time variable, respectively. In our context, the field $m = m(x, t)$ represent the polymer concentration while $\phi = \phi(x, t)$ is related to solvent concentration such that in our ternary mixture, solvent concentration is given by $c(x, t) := 1 - \phi(x, t)$ for all $(x, t) \in \bar{\Omega} \times [0, T]$, including thus the initial solvent concentration, denoted c_0 . Finally, the operator \star represents the standard convolution operator. For more details, see [13]

Note that (1.4) is a coupled parabolic system with nonlinear, nonlocal and degenerate drift, which raise a couple of mathematical challenges that we will point out in paper I and II.

1.4 Goal of the thesis

The overarching goal of this thesis is to investigate, from a mathematical perspective, the three dimensional morphology formation of interacting ternary mixtures as it applies to fabrication of organic solar cells. This goal is lofty and

entirely unreasonable for the short period of time allotted, however, on the way towards this lofty goal many questions demand an answer.

The models presented in section 1.1 are interesting by themselves but together they provide a richer picture of the modeled situation. The lattice model lets us control the microscopic dynamics whilst the continuum model is far more amenable to be extended (to include, for instance, non-equilibrium boundary effects like solvent evaporation). The continuum model is a hydrodynamic limit of the Blume-Capel model as derived in [13]. Since the probabilistic derivation involve the introduction of nonlocal interactions, it is clear that some information of the microstructure is lost during the derivation of the continuum model. In [5, 12], using the Blume-Capel model and its hydrodynamic limit, the morphology formation, growth rates and domain sizes were studied in the two dimensions. The two dimensional studies revealed distinct solvent concentration regimes were the types of morphologies obtained change drastically. We have in mind the following basic questions:

- Q₁. Can our models capture morphology formation in three space dimensions ? Under what conditions can the models capture the aforementioned morphologies ?*
- Q₂. Do the 3D models exhibit distinct types of morphologies for different solvent concentrations ?*
- Q₃. Is the proposed numerical scheme suitable ? It is mass conservative? Does it properly dissipate energy? Is it well-posed? Can we show convergence rates ? Is it stable with respect to parameters and initial conditions ?*

Beyond these mathematical questions there are several challenges from a computational point of view:

- C₁. Implementing a memory efficient Newton solver for large grids*
- C₂. Visualizing morphologies in three space dimensions*
- C₃. Characterize properties like connectivity, typical size, interface area, distance to boundary for the obtained morphologies*

1.5 A personal story behind computing in 3D the approximate solution to system presented in (1.4)-(1.5)

In this section, I would like to tell a few things about the philosophy behind my computational work, how I think about it and, in the process, highlight certain computational aspects of the continuum model which proved more complicated than expected.

Mathematical analysis of partial differential equations (PDEs), in my view, primarily concerns itself with the study of qualitative questions. In essence, what questions actually make sense to ask. In concrete language, does my model have a unique solution? In which space it exists? What properties can be deduced *a priori*? Numerical analysis of PDEs is slightly more concrete, its purpose is to describe how one can find a good approximation to the solution of the model at hand. Scientific computing in the computational mathematics framework is getting *de facto* to the approximate solution.

One might suspect that because both scientific computing and computational mathematics are primarily about the very concrete task of getting to the approximate solution it is the most straightforward. But, the concreteness of the task also brings forth practical problems to face.

I started my work from previous studies on a two dimensional (2D) implementation written in Python, with the goal to extend it to three space dimensions (3D). A simulation of a complex problem in 2D is nothing to scoff at but, when the physical system you model is indeed in 3D, a 2D simulation can only provide a partial view. In mathematical analysis context, this is usually described as a straightforward task eventually involving adaptations at the level of Sobolev embeddings, but on the computational side, this involves detailed considerations that simply is not necessary in two space dimensions.

The first thing is that the degrees of freedom in the discrete model will multiply. The three dimensional simulations will thus greatly increase the computational cost (time and resources). Numerically, such extensions may also change the stability and behavior (geometry of morphologies, connectivity, local curvature) of the obtained approximations, which simply tells that complexity increases with the increase of space dimension.

The main issues with the model we are investigating involve a difficult interplay between the nonlinearity and the nonlocality present in the drift. In the 2D case, the model was linearized and solved by using an explicit Runge-Kutta scheme while the convolution was handled in Fourier space. The first version of our 3D implementation was written in Python and utilized the same strategy. The obtained approximation showed either numerical instabilities or computational costs that rendered any realistic exploration of the model impossible. This happened because the extra space dimension changed the CFL condition. The timestep required to maintain numerical stability decreased substantially. The obvious answer to this issue is to switch the time integration to an implicit scheme, which in turn means that one needs a different strategy to handle the nonlinear terms. We decided to implement an iterative Newton method to handle the nonlinear drift. In doing so, one typically constructs the Jacobian matrix which for a system like ours becomes very large. This construction was too large to handle on any generic

laptop. Luckily, we had access to computational resources through the National Academic Infrastructure for Supercomputing in Sweden (NAISS). This allowed us to solve our problem on a 64^3 cubic grid. The results looked very grainy and unable to properly resolve the interfaces between phases. The results were also a far cry from comparing the continuum model to the resolution available to us from the stochastic Blume-Capel simulations that was computed on a 256^3 lattice. At this point, I decided to switch the coding language to `Julia`, a language built around high performance computing. This sped up the simulations but it still did not allow us to increase the resolution.

To tackle the resolution problem I implemented a matrix free version of the Newton scheme, meaning that we never had to construct the Jacobian matrix. Instead, we compute only what the Jacobian's effect would be on a vector. This drastically reduced the amount of random access memory (RAM) needed for the simulation. Through all these adjustments and re-implementations, the typical simulation time (of course dependent on model parameters) was on the scale of hours using resources from NAISS. This finally allowed us to compute our system on a 256^3 cubic grid so that qualitative comparison with the stochastic simulations were possible, and for the time being we were happy with the result.

Extensions to the computations presented within this thesis, the implementation and subsequent simulation for our model with non-equilibrium boundary evaporation of the solvent have also been performed. This seemingly small extension led to a complete rewrite of the `Julia` implementation such that it now runs on a Graphical Processing Unit (GPU) as opposed to the traditional Central Processing Unit (CPU). This became necessary because the introduction of evaporation in the model added a new timescale to the system, resulting in much longer simulation times.

List of publications

Papers included in the thesis

- (I) Cirillo, E. N. M., Jävergård, N., Lyons, R., Muntean, A., and Muntean, S. A. “3D morphology formation in a mixture of three differently averse components”. In: *Modelling and Simulation in Materials Science and Engineering* 33.055014 (2025)
- (II) Jävergård, N., Lyons, R., and Muntean, A. “Semi-discrete finite volume approximations of coupled evolution equations for ternary mixtures: Convergence and 3D morphological studies”. In: *Discrete and Continuous Dynamical Systems - S* 24 (2026), pp. 97–119

Other published work

Jävergård, N., Lyons, R., Forsmann, J., and Muntean, A. “Tunable correlation retention: A statistical method for generating synthetic data”. In: *Advances in Mathematical Sciences and Applications* 34.2 (2025), pp. 779–801

Jävergård, N., Nika, G., and Muntean, A. “Mathematics for energy systems: Methods, modeling strategies, and simulation”. In: *ROMAI Journal* 20.2 (2024), pp. 67–80

Preprints

Jävergård, N., Morale, D., Rui, G., Muntean, A., and Ugolini, S. *A hybrid model of sulphation reactions: stochastic particles in a random continuum environment*. 2025. arXiv: 2503.01856 [physics.chem-ph]. URL: <https://arxiv.org/abs/2503.01856>

Ounissi, O., Jävergård, N., and Muntean, A. *Orthogonal Procrustes problem preserves correlations in synthetic data*. 2025. arXiv: 2510.02405 [stat.ME]. URL: <https://arxiv.org/abs/2510.02405>

Author's contributions

This section clarifies the contributions that I, Nicklas Jävergård, have made to the manuscripts included in this thesis.

- I. **Cirillo, Emilio N M, Jävergård, Nicklas, Lyons, Rainey, Muntean, Adrian, and Muntean, Stela Andrea** 3D morphology formation in a mixture of three differently averse components, *Modelling and Simulation in Materials Science and Engineering*, 33.055014, (2025).

Own contributions (CRediT)¹:

Conceptualization, Investigation, Methodology, Software, Visualization, Writing – original draft, Writing – review & editing

Film formation from solvent evaporation in polymer ternary solutions is relevant for several technological applications, such as the fabrication of organic solar cells. The performance of the final device will strongly depend on the internal morphology of the obtained film, which, in turn, is affected by the processing conditions. We are interested in modeling morphology formation in 3D for ternary mixtures using both a lattice model and its continuous counterpart in the absence of evaporation. In our previous works, we found that, in 2D, both models predict the existence of two distinct regimes: (i) a low-solvent regime, characterized by two interpenetrated domains of the two polymers, and (ii) a high-solvent regime, where isolated polymer domains are dispersed in the solvent background. In the significantly more intriguing 3D case, we observe a comparable scenario both for the discrete and the continuous model. The lattice model reveals its ability to describe morphology formation even in the high solvent content 3D case, in which the three-dimensional nature of space could have prevented cluster formation. To realize the simulations we have written specific codes using the languages C and Julia. The codes closely follow the algorithmic dynamics governing the lattice and the continuum model.

¹<https://credit.niso.org/>

- II. **Jävergård, Nicklas, Lyons, Rainey, and Muntean, Adrian.** Semi-discrete finite volume approximations of coupled evolution equations for ternary mixtures: Convergence and 3D morphological studies *Discrete and Continuous Dynamical Systems - S*, 24 (2026) pp. 97-119

Own contributions (CRediT):

Conceptualization, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review & editing

Motivated by questions related to morphology formation in 3D involving interacting ternary mixtures, we propose a finite volume scheme to approximate numerically the unique weak solution to a coupled system of parabolic equations with nonlinear and nonlocal drift. The special feature of our system is that the coupling takes place precisely via the structure of the drift terms. We prove the convergence of the scheme towards the unique solution of the target evolution system and explore as well the stability of the solution with respect to selected parameters. We illustrate numerically in 3D the appearance of the wanted morphologies and compute as well the empirical order of convergence of the numerical approximations towards their limit.

Chapter 2

Results

2.1 Discussion of results in paper I

The results presented in paper I are of a qualitative nature. They highlight that both models are very well suited to describe and capture phase separation in three space dimensions.

We illustrate the capacity to simulate morphology formation by means of numerical simulations both for the Blume-Capel model and the continuum model for various parameter values. In Figure 2.1.1, we show the configuration¹ of the Blume-Capel model as well as its hydrodynamic limit. The two different polymer phases are given in yellow and blue whilst the solvent is displayed in red. As can be seen in Figure 2.1.1, both models show two distinct behaviors: one in the low solvent concentration regime and one in the high solvent concentration regime. In the low solvent concentration regime, our models show elongated regions for each phase with a thin layer of solvent at the separating interface. Forming morphologies that are similar to the bicontinuous structure typical to the two-state Ising model [6]. This observation is expected considering the nature of the Hamiltonian described for the Blume-Capel model in section 1.3.1 as well as the rescaled version used in the continuous counterpart presented in section 1.3.2. The qualitative appearance of the solutions between the two models remain similar as long as the scale factor γ (appearing in the continuum model) is small enough. In contrast to the low solvent concentration regime, in the high solvent concentration regime we observe spherical regions of each phase immersed in the ambient solvent for both models.

We also investigated how the domain sizes grow as a function of time. Such type of information was computed using the correlation function of the fields for different

¹We refer to "configuration" as the spatial distribution of the occupant species (continuous or discrete)

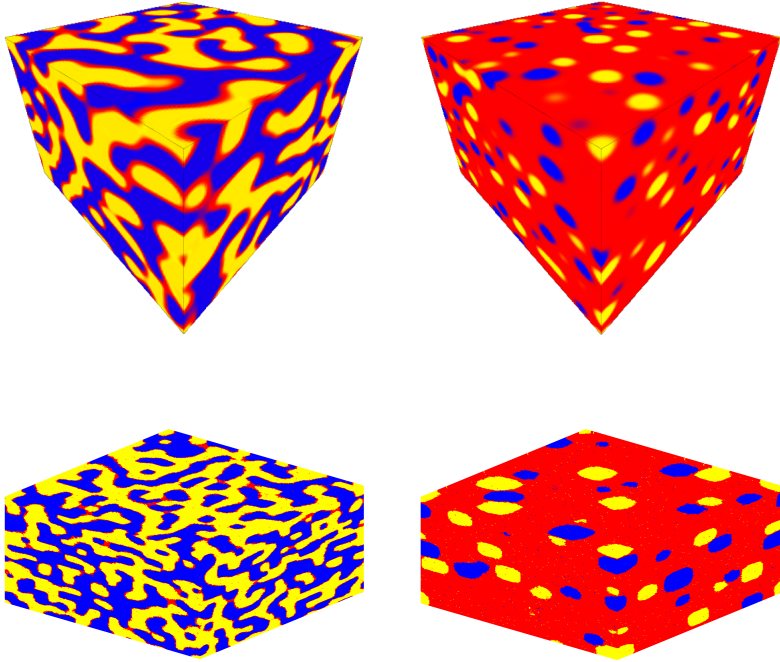


Figure 2.1.1: In the top row we show the configuration obtained using the continuum model with solvent concentration $c_0 = 0.2$ (left) and $c_0 = 0.8$ (right). In the bottom row we show the configuration obtained using the Blume-Capel model on 128^3 lattice with solvent concentration $c_0 = 0.2$ (left) and $c_0 = 0.8$ (right).

solvent concentration for the two models as can be seen in Figure 2.1.2, restated here from Figure 14 paper I. In this figure we see that the growth exponent for the stochastic model is $1/3$ and appears to be stable for different solvent concentrations. We observe the same phenomenon in the continuum model. However here, the exponent is slightly lower with a value of $1/4$.

2.2 Discussion of results in paper II

In paper II we propose a semi-discrete finite-volume scheme for approximating the weak solution the continuum model presented in paper I. We prove key results establishing the convergence of the semi-discrete finite-volume scheme. We also show the effect of different values of solvent concentration as well as the inverse temperature β on the model output.

In order to properly discuss the results in paper II, we start by presenting the semi-discrete finite-volume scheme, for more details we refer the reader to section 2.3 and section 2.4 of paper II. We denote our finite-volume approximation as (m_h, ϕ_h) and their accompanying space of grid functions as $\mathcal{F}_h(\Omega_h)$, where Ω_h is a uniform

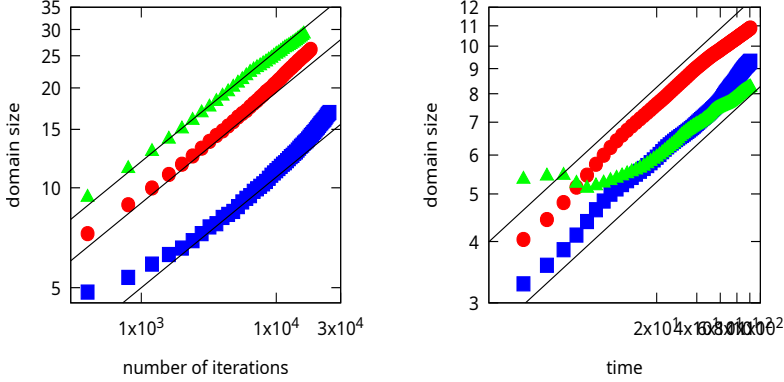


Figure 2.1.2: Left panel: loglog plot of the spherical average of the domain size versus time for the discrete space model with $L = 128$, $c_{+1} = c_{-1} = (1 - c_0)/2$, and $c_0 = 0.1$ (blue squares), 0.4 (red disks), and 0.8 (green triangles). The three straight lines have equations $0.50t^{1/3}$, $0.90t^{1/3}$, and $1.20t^{1/3}$. Right panel: analogous result for the continuum model for $\phi_0 = 0.1, 0.4, 0.8$ (analogous color code). The two straight lines are $2.5t^{1/4}$ and $3.9t^{1/4}$. This figure is taken from [4], Figure 14.

Cartesian grid on Ω , with grid size h .

For all $t \in (0, T)$ and for any $h > 0$, find the pair $(m_h(t), \phi_h(t))$ satisfying the equations

$$\begin{cases} \frac{d}{dt} m_h = \Delta_h m_h - \operatorname{div}_h(2\beta(\phi_h - m_h^2) \nabla_h J_h \otimes m_h), & \text{in } \Omega_h \times (0, T), \\ \frac{d}{dt} \phi_h = \Delta_h \phi_h - \operatorname{div}_h(2\beta m_h(1 - \phi_h) \nabla_h J_h \otimes m_h), & \text{in } \Omega_h \times (0, T), \\ m_h, \phi_h \text{ periodic at } \partial\Omega_h \times (0, T), \\ m_h(0) = m_{0h}, \quad \phi_h(0) = \phi_{0h}, & \text{in } \bar{\Omega}_h, \end{cases} \quad (2.1)$$

where ∇_h , div_h , and Δ_h denote the discrete counterparts of the gradient, divergence, and Laplacian, respectively. And where \otimes is defined as

$$(f_h \otimes g_h)_{i,j,k} := h^3 \sum_{r=0}^N \sum_{s=0}^N \sum_{t=0}^N f_{r,s,t} g_{i-r,j-s,k-t},$$

for two grid functions $f_h, g_h \in \mathcal{F}_h(\Omega_h)$ where the indices $i, j, k \in \{1, 2, \dots, N\}$ refer to evaluation on the nodes $(x_i, y_j, z_k) \in \Omega_h$. The system (2.1) comprises of coupled, nonlinear, ordinary differential equations that are locally well-posed in the sense that there is some time $T' \in (0, T)$ such that

$$(m_h, \phi_h) \in [C^1(0, T')^N \cap C[0, T']^N]^2$$

satisfies (2.1) with $m_h(0) = m_{0h}$ and $\phi_h(0) = \phi_{0h}$, where m_{0h} and ϕ_{0h} are suitable

approximations of m_0 and ϕ_0 , cf. (1.5). Here $\mathcal{N} := N^3$ is the size of the spatial grid used for the finite volume approximation. Now, we define the weak form of this system.

Definition 2.2.1. *We say that the pair $(m_h, \phi_h) \in C^1(0, T; \mathcal{F}_h(\Omega_h))^2$ is a weak solution to (2.1) if*

$$\begin{cases} \left(\frac{d}{dt} m_h, \eta_h \right)_{\mathcal{L}^2(\Omega_h)} + (\nabla_h m_h, \nabla_h \eta_h)_{\mathcal{L}^2(\Omega_h)} = (2\beta(\phi_h - m_h^2) \nabla_h J_h \otimes m_h, \nabla_h \eta_h)_{\mathcal{L}^2(\Omega_h)}, \\ \left(\frac{d}{dt} \phi_h, \psi_h \right)_{\mathcal{L}^2(\Omega_h)} + (\nabla_h \phi_h, \nabla_h \psi_h)_{\mathcal{L}^2(\Omega_h)} = (2\beta m_h (1 - \phi_h) \nabla_h J_h \otimes m_h, \nabla_h \psi_h)_{\mathcal{L}^2(\Omega_h)}, \\ m_h(0) = m_{0h}, \quad \phi_h(0) = \phi_{0h}, \end{cases} \quad (2.2)$$

for all $\eta_h, \psi_h \in \mathcal{F}_h(\Omega_h)$.

Throughout our analysis, we make use of the following standing assumption

$$0 \leq |m_0| \leq \phi_0 \leq 1 \text{ almost everywhere in } \bar{\Omega}, \quad (\text{H})$$

and of its discrete analog

$$0 \leq m_{0h} \leq \phi_{0h} \leq 1 \text{ in } \bar{\Omega}_h. \quad (2.3)$$

The structure of the proof of strong convergence of the finite-volume approximates to the weak solution of (2.2) is quite straightforward and relies on compactness arguments. To this aim, we apply Aubin-Lions Compactness Theorem, restated here.

Theorem 2.2.1 (Aubin–Lions Compactness Theorem). *Let $B_0 \hookrightarrow B \hookrightarrow B_1$ be Banach spaces with compact embedding $B_0 \hookrightarrow B$ and continuous embedding $B \hookrightarrow B_1$. If the sequence $\{u_n\}$ is bounded in $L^p(0, T; B_0)$ and the sequence $\{\partial_t u_n\}$ is bounded in $L^q(0, T; B_1)$ (with $1 < p, q < \infty$), then $\{u_n\}$ is relatively compact in $L^p(0, T; B)$.*

Looking at the hypothesis of the Aubin–Lions Compactness Theorem, we need show that the linear interpolation of our solutions (m_h, ϕ_h) belong to $L^2(0, T; H_{\sharp}^1(\Omega))$ and show that their time derivatives $(\partial_t m_h, \partial_t \phi_h)$ lie in $L^2(0, T; L_{\sharp}^2(\Omega))$ and thus also in $L^2(0, T; H_{\sharp}^{-1}(\Omega))$. These properties are proven in Lemma 2.2.2 and Lemma 2.2.3, restated below. We select the spaces $B_0 := H_{\sharp}^1(\Omega)$, $B := L_{\sharp}^2(\Omega)$ and $B_1 := H_{\sharp}^{-1}(\Omega)$. It then follows that the collections of our sequences of finite-volume approximations, are relatively compact in $L_{\sharp}^2(\Omega)$.

Lemma 2.2.1. *Let m_h, ϕ_h be solutions to (2.1) such that*

$$|m_{0h}(x_h)| \leq \phi_{0h}(x_h) \leq 1$$

for all $x_h \in \Omega_h$. Then it holds

$$|m_h(x_h, t)| \leq \phi_h(x_h, t) \leq 1$$

for all $t \in [0, T]$ and for all $x_h \in \Omega_h$.

Lemma 2.2.2. *If (H) holds, then there exists a constant $C^* > 0$ independent of h such that*

$$\max_{t \in [0, T]} \left(\|m_h\|_{\ell^2(\Omega_h)}^2 + \|\phi_h\|_{\ell^2(\Omega_h)}^2 \right) \leq C^*$$

and

$$\int_0^T \left(\|\nabla_h m_h\|_{\ell^2(\Omega_h)}^2 + \|\nabla_h \phi_h\|_{\ell^2(\Omega_h)}^2 \right) ds \leq C^*.$$

Lemma 2.2.3. *If (H) holds together with $\|\dot{w}(0)\|_{\ell^2(\Omega_h)}^2 + \|\dot{v}(0)\|_{\ell^2(\Omega_h)}^2 \leq C$ for some $C > 0$ independent of h , then there exists a constant $C^* > 0$ independent as well of h such that*

$$\max_{t \in [0, T]} \left(\left\| \frac{d}{dt} m_h \right\|_{\ell^2(\Omega_h)}^2 + \left\| \frac{d}{dt} \phi_h \right\|_{\ell^2(\Omega_h)}^2 \right) \leq C^*.$$

The above Lemmas allow us to prove the following result, which shows that the linear interpolations of $(m_h, \phi_h) \in L^2(0, T; \mathcal{F}_h(\Omega_h))^2$ converge strongly to the solution of (2.2), which is the main result in paper II.

Theorem 2.2.2. *Assume the hypothesis of Lemma 2.2.3 to hold true and take $(m_h, \phi_h) \in C(0, T; \mathcal{F}_h(\Omega_h))^2$ to be a solution in the sense of Definition 2.2.1. Let $\hat{m}_h, \hat{\phi}_h$ be the linear interpolates of the solutions to the semi-discrete scheme (2.2). Then, as $h \rightarrow 0$, there exists subsequences \hat{m}_h and $\hat{\phi}_h$ such that*

$$\hat{m}_h \rightarrow m, \quad \hat{\phi}_h \rightarrow \phi \quad \text{strongly in } L^2(0, T; L^2_{\#}(\Omega)),$$

where (m, ϕ) is the unique weak solution of (2.2).

In this paper we also prove a stability estimate of the obtained solution with respect to the initial conditions and to the inverse temperature parameter β in (2.4), see section 3 of paper II [in particular Theorem 3.1] for details. Furthermore, we estimate via numerical experiments the convergence rate of the used approximation scheme, viz. there exist $c_1 \geq 0, c_2 \geq 0$ independent of h such that

$$\begin{aligned} & \|m_1(t) - m_2(t)\|_{L^2(\Omega)}^2 + \|\phi_1(t) - \phi_2(t)\|_{L^2(\Omega)}^2 \leq \\ & \leq \left(\|m_1(0) - m_2(0)\|_{L^2(\Omega)}^2 + \|\phi_1(0) - \phi_2(0)\|_{L^2(\Omega)}^2 \right) e^{C_2 t} \\ & + \frac{C_1}{2C_2} |\beta_1 - \beta_2|^2 \left(e^{C_2 t} - 1 \right), \end{aligned} \tag{2.4}$$

holds for all $t \in (0, T)$, as well as

$$\begin{aligned} & \|m_{1h}(t) - m_{2h}(t)\|_{\ell^2(\Omega_h)}^2 + \|\phi_{1h}(t) - \phi_{2h}(t)\|_{\ell^2(\Omega_h)}^2 \leq \\ & \leq \left(\|m_{1h}(0) - m_{2h}(0)\|_{\ell^2(\Omega)}^2 + \|\phi_{1h}(0) - \phi_{2h}(0)\|_{\ell^2(\Omega)}^2 \right) e^{C_2 t} \\ & + \frac{C_1}{2C_2} |\beta_1 - \beta_2|^2 \left(e^{C_2 t} - 1 \right). \end{aligned}$$

The grid functions $m_{1h}(t), m_{2h}(t), \phi_{1h}(t), \phi_{2h}(t) \in \mathcal{F}_h(\Omega_h)$ are the discrete versions of m_1, m_2, ϕ_1, ϕ_2 and $\|\cdot\|_{\ell^2(\Omega_h)}$ denotes the discrete L^2 norm.

Chapter 3

Outlook

In this licentiate thesis, we have studied the morphology formation in interacting ternary mixtures. This topic is important from an application point of view in the context of organic solar cells and is rich with mathematical features, not yet fully explored. In this closing chapter, we list a couple of open questions and extensions that could be addressed in future work.

1. The geometrical properties of the obtained three dimensional morphologies (see paper I) require careful investigation to complement the information already extracted regarding growth rates and domain sizes. We expect that tools from differential geometry will prove useful, for instance, in computing local curvature information to quantify the connectivity of each phase.
2. The finite volume scheme presented in paper II has been analyzed and proven to converge for a semi-discrete setting. The natural next step is to investigate the convergence properties of the fully discrete counterpart of our scheme.
3. Boundary evaporation is an important mechanism in the laboratory experiments producing the morphologies of organic solar cells. As we wish to describe in mathematical terms the laboratory situation, our model must be extended to include evaporation. Such an extension would require us to adapt both the theoretical and algorithmic frameworks to deal with our system of partial differential equations with nonlocal drift whilst interacting with nonperiodic boundary information.
4. An alternative to using Newton iterates to approximate the solution to our system of partial differential equations with nonlocal drift is to make use of spectral methods. We believe this strategy would speed up computations by a substantial margin, helping massively our investigation of parameter regimes and interface structure at smaller scales.
5. The current structure of our target system of partial differential equations

does not carry information of how strong the mixture components interact with each other. In the physical system, we have two very different polymer species immersed in a solvent - each of the components exhibits distinct affinities to one another. It would be interesting to use molecular dynamics simulations to obtain interaction parameters between two polymer species in different solvent concentration regimes and then introduce them to our continuum model. This way our model becomes naturally more interpretable as it would better describe the physics of the situation.



A finite volume scheme for a conservative hydrodynamic limit of the Kac-Blume-Capel model

Morphology formation in thin films produced from a ternary solution is crucial for the performance of organic solar cells. Both the separation of excitons into free charges as well as the charge transport that follows depend on the shape and connectivity of the distinct polymer regions (the morphology). In this thesis, we study morphology formation from two different perspectives: A lattice-based Blume-Capel model with Kawasaki dynamics, and then a continuum system of coupled parabolic equations with nonlinear and nonlocal drift. The objective of this licentiate thesis is to represent morphology formation in three space dimensions using these two models. We relate our work to previous two-dimensional results for different parameter regimes. At the technical level, we construct a semi-discrete finite volume scheme to approximate the weak solution of our continuum model and implement it in Julia. We prove a convergence result of our semi-discrete scheme as well as a stability result of the weak solution with respect to temperature variations - a key parameter in the model. Looking at both the lattice model and the continuum parabolic system, we quantify and compare growth rates of the formed domains. Finally, we perform numerical experiments confirming convergence of our scheme and the effect of parameters on the obtained solution. These results provide a solid foundation for future extensions, including the evaporation of a mixture component.

ISBN 978-91-7867-690-3 (print)

ISBN 978-91-7867-691-0 (pdf)

ISSN 1403-8099

LICENTIATE THESIS | Karlstad University Studies | 2026:21
