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REVIEWS

Applications of the Phase-Field Method for the Solidification of Microstructures in Multi-Component Systems

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Abstract | The solidification of multicomponent alloys is of high technical and scientific importance. In this review, we describe the ongoing research of the phase-field method for the solidification of dendritic and eutectic structures. Therefore, the corresponding experimental and theoretical investigations are presented. First, an overview of the historical development in solidification research is given. Thereafter, the ongoing progress of the phase-field models is reviewed. Then, we address the experimental and simulative investigations of different forms of dendritic and eutectic solidification. We distinguish between thermal and solutal dendritic growth as well as thin-sample and Bridgman furnace experiments of eutectic growth. Impurity-driven Mullins-Sekerka instabilities like cell structures, eutectic colonies and spiral dendritic growth are presented. Then, validation methods for the comparison between simulations, experiments and theoretical approaches are addressed. Subsequently, related aspects to simulate solidification are introduced. Especially, further physical aspects and computational optimizations are considered. Concluding, possible future research in the context of the phase-field method for solidification is discussed.

1 Introduction

Economical and ecological challenges demand the development of new materials. The application of novel and optimized materials requires defined properties, which are strongly influenced by the characteristics of their chemical elements and composition as well as by the evolving microstructure as a result of the particular manufacturing processes.[1–3] One important forming process with high technical relevance is solidification. The possibility of exploiting the advantages of different chemical components is in focus of the research for new materials as they are needed in applications with high mechanical and thermal requirements, e.g. in automotive industry, aerospace, turbines and in lead-free solders.[4,5] For example, by adding different additive components like Cr, Mo, W, C, Nb to unary metals or binary alloys, the creep resistance at high temperatures, the fracture toughness or the stiffness can be significantly improved. The periodic table of elements includes 80 metallic components, which have the potential to form 82,160 different ternary alloys. From these, around 3000 systems have partially been investigated until now. For higher-ordered systems, like quaternary systems, the number of possible alloys increases enormously.

Since the Bronze Age, humans combine different materials in solidification experiments. First analytical models to describe the solidification processes were established in the 1940s.[6] Fundamental work for the understanding of solidification processes were conducted by Mullins and Sekerka,[7–9] performing stability analysis, and

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Jackson and Hunt,[10] studying the eutectic pattern formation. In 1986, Kurz and Fisher summarize previous theories in their book *Fundamentals of Solidification*.[11] The following improvements in theories and experiments for the understanding of solidification processes of dendritic and eutectic growth are collected in.[12–16]

Caused by the exponential increase of computational power,[17] simulations continuously gain importance for the development of new materials, besides the classical fields of experiment and theory.[6] Especially for the simulation of microstructure evolution processes, the phase-field method (PFM) has emerged as a powerful and versatile technique in the recent years.[18] Several authors show the accordance between the PFM and sharp-interface theories.[19–26] The applicability for simulating dynamic processes is demonstrated in.[27] The progress in computer science is reflected in increasing simulation domains and the more detailed phase-field models leading to a better understanding of solidification. Figure 1 shows an exemplary selection of simulations to demonstrate the methodological process from two-dimensional to three-dimensional pattern formations as well as from unary to ternary alloy systems.

In the PFM, the boundaries between two or more phases are described as mathematically steady transitions. This allows the phase-field method to simulate the microstructure evolution without interface tracking, in contrast to the classical finite element method (FEM) for structural mechanics. Compared with first principle methods, the PFM enables the calculation of larger physical domains, through the different used length and time scales. Therefore, the PFM bridges the gap between the

Sharp-interphase: A jumping discontinuity as the transition between two phases.

Interface tracking: Repeated meshing of the phase boundaries in the domain to follow the evolution of the interface.

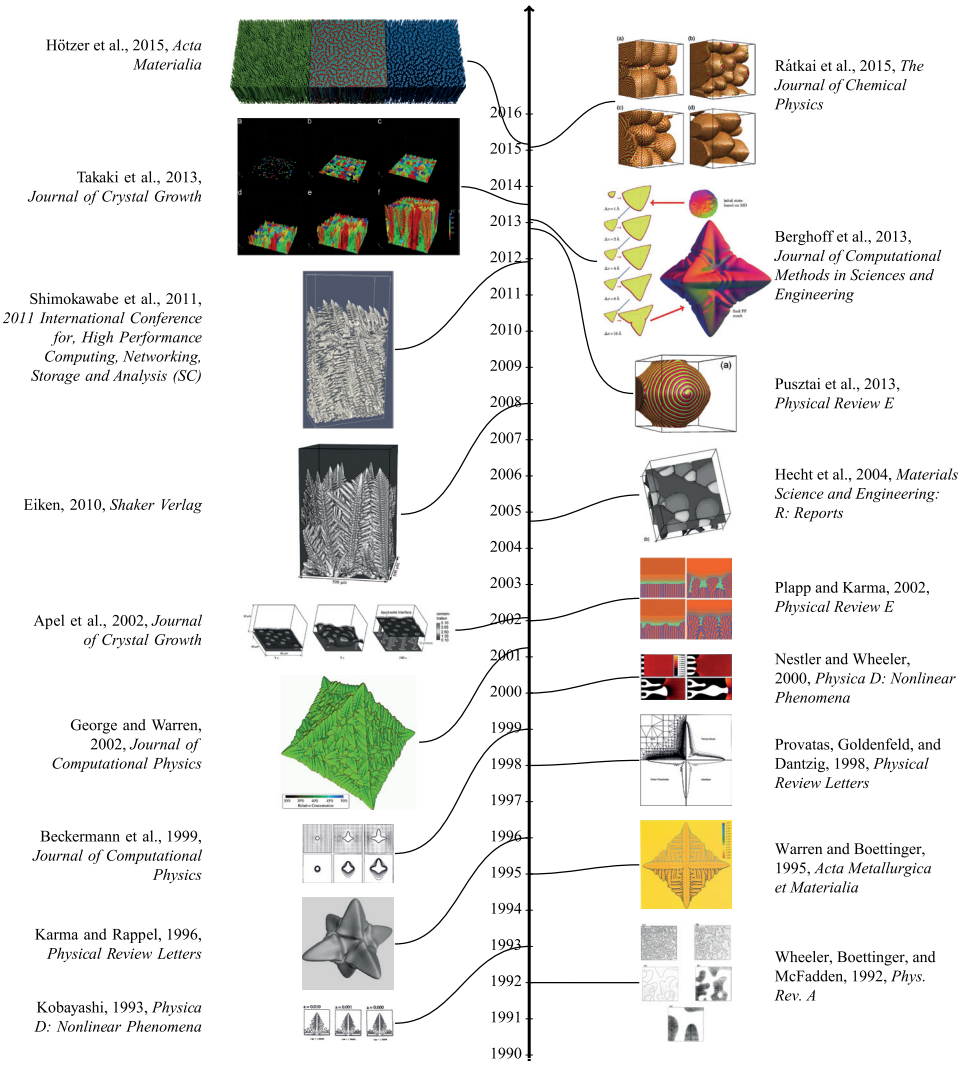


Figure 1: Phase-field simulations of solidification microstructures from the 1990s until 2015 showing the increasing complexity from two-phase one-component systems in 2D to multiphase multicomponent systems in 3D as the current state of art.

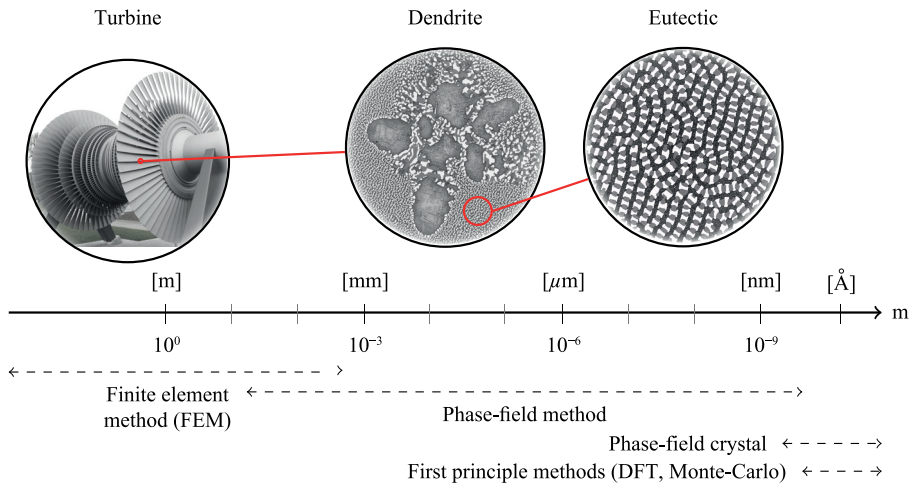


Figure 2: Overview of the typically applied simulation methods for different length scales.

atomistic scale and the macroscopic scale, as illustrated in Fig. 2. An overview of the PFM and its advantages and disadvantages is summarized by Qin and Bhadeshia.[28]

The developments and the current research in the field of PFM are collected in the review articles.[18,28–42] A detailed description of the phase-field method is given by Provatas and Elder,[43] and the fundamental physical principles are explained in.[6,11,15,44,45]

2 Development of the Phase-Field Method

In the following section, an overview of the development of the phase-field method (PFM) is given. For this purpose, the fundamental principles are introduced and the derivation of the driving forces for solidification, and the modifications of the models are described.

The concept to interpret the interface between two phases as a diffuse area is introduced by van der Waals in the 1870s,[46] and is extended with an order parameter as the Ginzburg-Landau functional theory in the 1950s.[47,48] Cahn and Hilliard[49–52] as well as Allen and Cahn[53] explain the derivation of partial differential equations from the energy functionals of Ginzburg-Landau type through the calculus of variation, leading to the evolution equations of the order parameters. The evolution of the order parameters and following the transformation from one phase to another, is driven by the energy minimization of the total system.

During the ongoing development of the PFM, different thermodynamic potentials are proposed and used as driving forces for the phase transition. One approach is based on the minimization of the Helmholtz free energy,[54–61] whereas a different development line applies, the maximization of entropy,[62–65] which is introduced by Penrose and Fife.[62] Another approach is based on the minimization of the Grand potential difference. During the phase transition under near equilibrium conditions, the chemical potentials of the phases are equal,[66,67] which has been exploited by several phase-field models.[68–70] From, this the driving force for the phase transition can be described by the difference of the Grand potentials.[71–74] The advantage of the Grand potential formalism is the decoupling of the interface energies and the interface thickness, caused by the usage of the projection of the thermodynamic energies instead of the energies itself.[72]

Thermodynamic consistent models are derived, based on the aforementioned thermodynamic potentials, in.[62,75–77] Other multiphase-field models are proposed by Steinbach et al.,[78] Garcke et al.[23] as well as Nestler et al.,[79] allowing to simulate the interaction of more than two different phases. As further extension of multiphase-field models, multicomponent models for solidification are introduced in.[64,80–83] After extending the PFM to multiple phases and multiple components,[80] Garcke et al. compare their approach to[62,84] and classical sharp interface models for validation, and demonstrate the equivalence in special cases.

The coupling of a multicomponent multiphase-field model with thermodynamic databases (CALPHAD) using a linear extrapolation scheme, is suggested by Eiken[1] et al.[83] and reproduces the diffusion equation of[67] and,[82] without the restriction to dilute solutions from earlier works of their group.[22]

[1]Changed from J. Tiaden to J. Eiken.

The more complex models and the increasing number of equations to simulate multicomponent and multiphase systems in representative volume elements, require a higher computational effort. The necessity of representative volume elements for the simulation of ternary eutectics is demonstrated in[85] and guidelines to assess these are presented in.[74]

To reduce this effort, various optimizations of the models are introduced. Karma et al.[86,87] introduce a mathematical optimization to reduce the computational effort, due to a smaller ratio between capillary length and interface thickness. To fix the number of order parameters to a constant value in multiphase systems, Kim et al.[88] exploit the limited number of nearest neighbours, of each phase, which is further adapted in a variation in.[89,90]

Due to the artificially enlarged interface in the PFM and the deviation from sharp interface models, a solute trapping effect occurs. This solute trapping effect is studied by Wheeler et al.[91] and Boettinger et al.[92] as well as several other authors.[93–95] To balance the non-physical effect, a so-called *anti-trapping-current* is proposed,[96–99] which however increases the computational effort.

Solute trapping effect: Artificial solute flow in the interface, due to a deviation of the phase-field interface profile from the sharp-interface limit.

Anti-trapping-current: Artificial parameter in phase-field models to balance the solute trapping effects.

Different coupling approaches of thermodynamic CALPHAD databases with phase-field models[100–109] are exploited to study the microstructure evolution of real systems.[110–112] An overview of different phase-field models is given by Caginalp and Xie,[59] Almgren[113] and Sekerka.[16]

3 Solidification Morphologies

Different applications of phase-field models are discussed to simulate solidification, the process describing the phase transition from liquid to solid. Thermodynamically, a phase transition can be controlled by concentration, temperature and pressure. In the phase-field models for solidification, the pressure is usually assumed to be constant, and hence the process only depends on concentration and temperature. To study directional solidification, the growth direction and the velocity is controlled by an imprinted temperature gradient. Experimentally, directional solidification is investigated by thin sample and Bridgman furnace experiments.

Depending on the process conditions during solidification and on the chemical elements, various transition mechanisms accompanied by characteristic microstructures occur, like dendrites, eutectics and peritectics as well as spinodal decomposition. An overview of peritectic solidification can be found in the work of Akamatsu and Plapp.[114] Spinodal decomposition is extensively studied in the publications.[115–120] The present review embraces different forms of dendritic and eutectic solidification. First, we focus on dendritic growth of two phases with one or two components. Thereafter, the solidification of multicomponent and multiphase systems for eutectic and off-eutectic alloys is reviewed.

3.1 *Dendritic growth*

Tree-like structures, as they can be found in many solidified samples, are labeled as dendrites. The word originates from the Greek term déndron, meaning tree. The first mathematical model to describe dendritic growth is formulated by Ivantsov et al. in 1947,[121] which is often used for the validation of phase-field models. Simulating dendritic growth can be distinguished between thermal and solutal growth as well as a combination of both. In thermal dendritic growth processes, the arising of different tips from the nuclei or existing dendrite arms is caused by fluctuations in the temperature fields. Lively investigations are documented in the 1990s.[19,24,57,86,87,122–127] One of the first 2D simulations of thermal dendritic growth is reported by Kobayashi in 1993,[57] presenting isotropic and anisotropic growth. Depending on the anisotropy modes and the employed noise, up to three side branches evolve.

Karma and Rappel present a computationally enhanced phase-field model for calculating dendritic solidification of pure materials by reducing the capillary length,[86] and validate the results with the sharp interface limit. This model is used to simulate 3D dendrites in[123] and elaborate quantitative comparisons with experiments of succinonitrile in.[87]

Solutal dendritic growth is driven by fluctuations in the concentration field of the melt and was intensively studied in the 2000s.[82,96,103,128–142]

The ability to computationally analyze solutal dendritic growth in alloys with the PFM is demonstrated by Karma in 2001.[96] He presents an extended phase-field model to quantitatively study microstructural pattern formation in alloys with a realistic solid diffusivity.

Warren and Boettinger[143] suggest a coupling method for thermal and solutal solidification and assume a negligible heat transfer due to computational reasons. In 2002, George and Warren[129] use the model of[143] to calculate one of the first large-scale simulations of solutal dendritic growth in a 3D domain of $500 \times 500 \times 500$ cells, performed on 32 CPUs.

In Fig. 3, two examples of directional competitive solutal dendritic growth are presented. To simulate the interplay of the various dendrite tips and their competition, large domains are required to resolve multiple dendrites. Fig. 3(a) shows the results of a $714 \times 714 \times 990$ cells domain, which is computed in 390 h using a single CPU in 2008 by Eiken.[144] A similar physical system is used by the Gordon-Bell Prize awarded work of Shimokawabe et al.[138] in 2011. In Fig. 3(b), their simulation with a domain size of $768 \times 1632 \times 3264$ voxel cells is depicted, calculated on the TSUBAME 2.0 supercomputer with 1156 GPUs. The success is continued two years later by Takaki et al.[141] in a $4096 \times 4104 \times 4096$ domain typically running for 2h 59 min. In 2016,[145] competitive dendritic and cell growth with large-scale two-dimensional phase-field simulations is investigated.

A combination of thermal and solutal dendritic solidification is introduced in[143] and investigated by Loginova et al.[146] and Emmerich et al.[147] Loginova et al. conclude that for low cooling rates and many nuclei an isothermal approach is applicable, but for higher cooling rates and fewer nuclei concentration, temperature evolution has to be considered.[146]

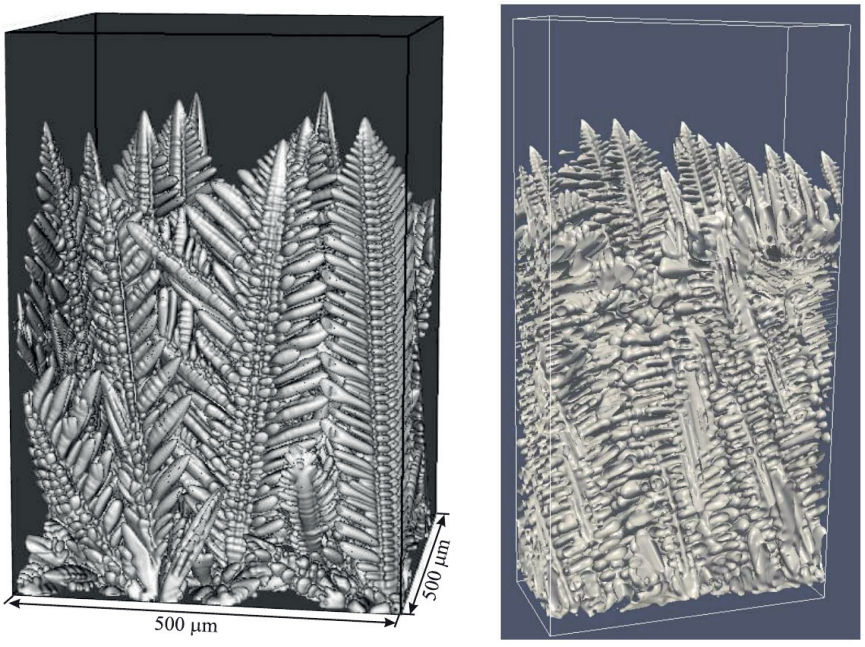
3.2 *Eutectic growth*

Beside solutal dendritic growth, simultaneous solidification of different distinct phases as eutectic solidification, can occur in systems with more than one component. According to[148] "*Eutectic solidification of a liquid is defined as a simultaneous precipitation of two or more phases via a eutectic reaction* $L \leftrightarrow \alpha + \beta$ *at constant temperature.*" The originally Greek term eutectic means *easy melting*. Classical eutectic structures arise at the eutectic point and slightly off-eutectic.[6]

Theoretical investigations of the stability of binary lamellar eutectic growth are derived in.[66,149–151] Different instability modes can occur like oscillation or tilted growth during directional solidification of eutectics. A growth is defined as tilted, if a stable deviation between the solid-solid interfaces and the imprinted growth direction from the temperature gradient exists. The phenomenon of tilt is theoretically studied by Kassner et al.[152] as well as the additional phenomenon of oscillation by Kassner et al.[153] and Karma and Sarkissian.[154]

Oscillation: Periodically repeating variations of the lamellar widths in a domain with a constant lamellar spacing.

For the experimental investigation of eutectic directional solidification, two kinds of methods have been established: Solidification of thin-film samples and Bridgman furnace experiments.



(a) Simulation of a 714 × 714 × 990 voxel domain on a single CPU for 390 h by Eiken[136,144] in 2008, using a highly optimized solver.

(b) Simulation of a 768 × 1632 × 3264 voxel domain using 1156 GPUs on the TSUBAME 2.0 by Shimokawabe et al.[138] in 2011.

Figure 3: Phase-field simulations of competitive directional dendritc growth with the formation dendrite network arrangements.

3.2.1 Thin-film growth and phase-field simulations of two-dimensional phenomena

The solidification of thin-film samples allows the in-situ study of organic transparent systems and the investigation of quasi two-dimensional solidification. This type of experiment provides insights into a mutual validation with theoretical approaches, which are primary derived for two dimensions, e.g. Jackson-Hunt[10] or Mullins-Sekerka.[7,8]

Especially, Faivre, Akamatsu, Bottin-Rousseau and co-workers investigate various binary organic systems using the thin-film sample technique.[155–162] A collection of the current research of in-situ observation of solidification patterns in diffusive conditions is presented by Akamatsu and Nguyen-Thi.[163] The research aims to carve out the influence of the processing conditions on the evolving lamellar arrangement and tilted growth of eutectics during directional solidification.

Caroli and Faivre[155] discuss the question of pattern selection in lamellar eutectic growth in 1995 and postulate that: "*experiments on thin-samples of lamellar eutectic do not support the idea of the existence of a selection principle leading to a unique final state*". Under specific conditions, the influence of growth and spatial boundary conditions acts as permanent sources of defects, resulting, e.g., in tilted or oscillating growth. Detailed studies of different aspects of tilted directional solidification are described in.[156,157] To identify the influence of anisotropy on the tilted growth, Akamatsu and co-workers[160–162] systematically vary the orientation of the solidification direction by the temperature gradient and determine a Wulff plot of the underlying anisotropy.[160]

Thin-sample experiments for ternary eutectics are reported by Rex.[164] It is found that slightly off-eutectic compositions in the system In-Bi-Sn lead to complex sequences of microstructure evolution.

To numerically explore thin sample-growth, two-dimensional phase-field simulations of binary eutectic systems are performed in.[20,70,79,165–167]

An accordance between the Jackson-Hunt approach[10] and phase-field simulations is revealed by Karma et al.,[20] Nestler and Wheeler[79] as well as Apel et al.[165] Simulations perform various instability modes such as different types of oscillations of the lamellar widths and varicose structures. A similar study for the system CBr_4-C_2Cl_6 is conducted by Folch and Plapp.[70]

Tilted growth, as experimentally reported in,[156,157,160–162] is simulated with the PFM by Ghosh and co-workers.[166–168] In these simulations as well as in the experiments, tilted growth is driven by interface energy anisotropies.

For ternary eutectic systems, further mechanisms can lead to tilted growth, as outlined in.[169,170] Hötzer et al.[170] show a relation between the interface energies and the tilt angle by systematical variation of the solid-solid and solid-liquid interface energy ratio in an idealized system. Ghosh et al.[166] and Hötzer et al.[170] discover a dependency of the tilt angle from the lamellar spacing and from the growth velocity. A concentration-driven tilt mechanism is described by Apel et al.[169] for Al-Ag-Cu. As further instability, various oscillation modes depending on the solid-solid interface anisotropy for ternary systems are discussed by Choudhury et al.,[171] continuing his previous work in[172] by breaking the symmetry conditions of the lamellae.

3.2.2 Bridgman furnace experiments and phase-field simulations of three-dimensional phenomenon

During the directional solidification in three dimensions, a wide range of different patterns can evolve. These patterns are of relevance for technical applications, due to their influence on the mechanical properties.[1–3] Experimentally, the directional solidification in three dimensions is realized by Bridgman furnace experiments.[173–175] Besides, eutectic structures also coupled eutectic and dendritic growth can be observed as exemplary shown in.[174]

Based on observations from experimental micrographs, Ruggiero and Rutter[176] predict five different patterns from geometrical considerations for the growth of ternary eutectics. These patterns are: (i) three lamellar phases, (ii) two lamellar phases and one fibrous phase, (iii) one lamellar phase and two fibrous phases, (iv) two fibrous phases in a continuous matrix phase and (v) three fibrous phases. Lewis et al.[177] graphically depicted the five patterns, which are reproduced in Fig. 4 (a) – (e). Corresponding simulations are found for the morphologies of (a) – (d)[178] and are shown in Fig. 4 (f) – (i).

A theoretical description of three different three-dimensional patterns by an extended Jackson-Hunt approach[10] is reported by Himemiya and Umeda.[179]

Visual comparison of micrographs from different ternary eutectic systems is conducted by Cooksey and Hellawell in 1967.[180] Further experimental investigations on the relation between lamellar spacing and growth velocity for different multicomponent systems are presented in.[173,174,181–183] The results are in accordance with the analytical Jackson-Hunt theory.[10]

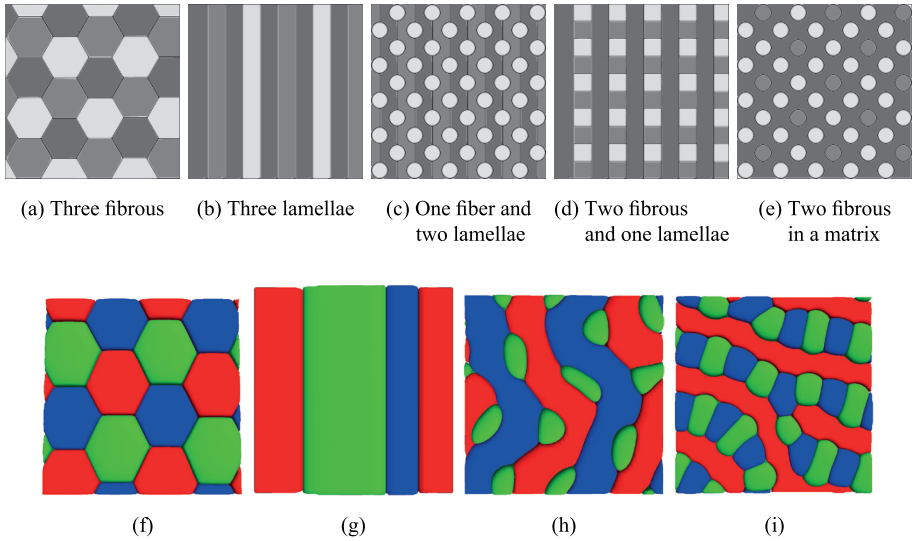


Figure 4: Theoretical patterns a–e parallel to the solidification front as given by[176,177] and phase-field simulation f–i as shown in.[178]

Ruggiero and Rutter consider the directional solidification of different ternary eutectic compositions consisting of the chemical elements Bi, In, Sn, Pb and Cd.[176,184–188] They refer to a wide range of patterns in their quenching experiments. Perrut and co-workers[189,190] elucidate long time in-situ observation of eutectic growth in three dimensions.

The ternary eutectic system Al-Ag-Cu is of growing scientific interest in the recent years. During the eutectic reaction, a wide range of patterns form.[180] Furthermore, the difference in the densities makes it favorable for low gravity experiments.[191] McCartney et al. treat the Al-Ag-Cu system in[192] to validate their theoretical work[150] on the stability of lamellar eutectics and achieve consistence to their predictions. Quenching experiments are conducted by Böyük and Marasli[175] and by Sargin[193] to study the pattern formation in longitudinal and transversal cross sections. The effect of convection and gravity on solutal configuration during univariant eutectic growth is investigated by Sargin.[193]

Genau and Ratke characterize the crystallographic orientation of the patterns with EBSD[194] and the phase arrangement with nearest neighbor statistics.[195] The statistical characterization is continued by Dennstedt and Ratke[196] using shape factors. In,[197] Dennstedt et al. compare experimental micrographs to phase-field simulations with polar plots. A first three-dimensional representation of the microstructure of the ternary eutectic system Al-Ag-Cu, obtained from synchrotron tomography, is depicted in[198,199] and in Fig. 5(b).

Other experimentally investigated ternary eutectic systems are, e.g. Nb-Al-Ni,[1] Al-Si-Ni[200] and Sn-Ag-Zn.[201]

To study Bridgman furnace experiments numerically with the PFM, two approaches are established. In the first approach, two-dimensional simulations of sections parallel to the solidification front are conducted. For these kind of simulations, nucleation has to be modeled to get comparable results. The second approach is to simulate the evolution in the total three-dimensional domain. The nuclei can be set as initial filling and patterns evolve from the nuclei during the solidification. An additional nucleation mechanism is not necessarily required. The advantage of the first approach is a multiple times smaller computational effort, compared with the second approach. But the 3D simulations allow to study the complex spatial interplay of different structures.

Two-dimensional phase-field simulations of Bridgman furnace experiments are, for example, executed in.[21,202–206]

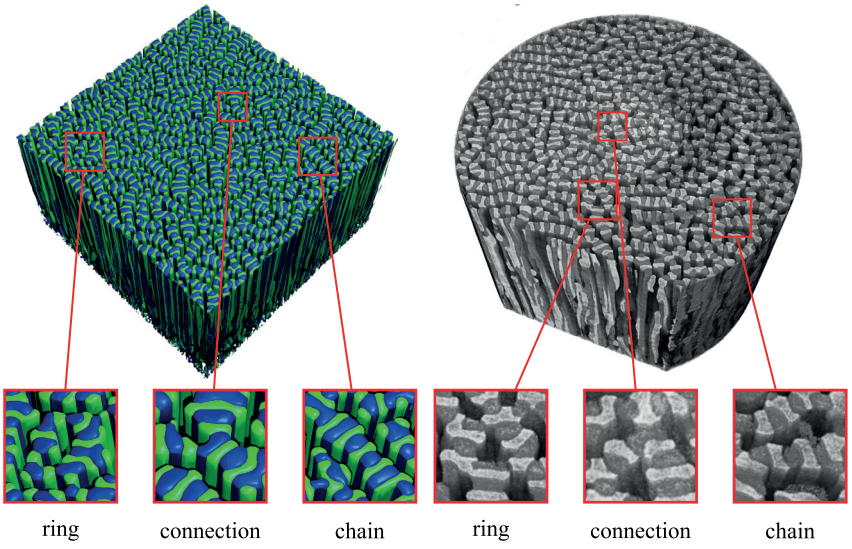
In 2002, Nestler and co-workers[203,204] employ a multiphase-field model to simulate eutectic growth in 2D, and illustrate a visual accordance for Al-Si. A similar approach is applied in 2015 by Kundin et al.[206] to study a combined peritectic-like and eutectic-like structures in the ternary system Al-Cu-Ni.

An overview of three-dimensional phase-field simulations of eutectic systems is given by Lewis et al.[207] in 2004 as well as Akamatsu and Plapp[114] in 2015.

Similar to the discussed oscillations and tilts in thin-film samples, further instabilities can occur in

Longitudinal cross section: A section parallel to the growth direction imprinted by the temperature gradient during directional solidification.

Transversal cross section: A section perpendicular to the growth direction imprinted by the temperature gradient during directional solidification.



(a) Large-scale phase-field simulation of the three-phase pattern formation in Al-Ag-Cu.

(b) Tomography reconstruction the experimental microstructure.

Figure 5: (a) Three dimensional simulation from Hötzer et al.[73] and (b) experimental results of directional solidification of the ternary eutectic system Al-Ag-Cu.[199] In the simulation and the experiment the matrix-phase is faded out.

3D. The stability of lamellar structures is embraced by Parisi and co-workers.[208–210] Disturbances of the concentrations and phase-fields, steady-state lamellar structures and arising instabilities are discussed.

A change of patterns is accompanied by a decrease of average front undercooling. A summary of the instabilities is given in.[211] Change of patterns caused by a transverse temperature gradient of binary systems, is investigated by Perrut et al.[212] The authors observed that a transverse temperature gradient leads to more aligned regular lamellar structures of the two arising phases instead of a labyrinth pattern. Ghosh and Plapp[167] extend their work of[166] for three dimensions and reveal a fundamental influence of the solid-solid interface anisotropy on the pattern selection depending on particular orientations.

Average front undercooling: Deviation of the average temperature at the solidification front from the solidus temperature for the considered melt concentration.

The three-dimensional growth of ternary eutectic directional solidification based on the model of[64] is studied by Choudhury et al.[172] Different patterns parallel to the solidification front are reported. With the approach presented in,[71,72] based on the minimization of the Grand potential difference, Choudhury[213] carved out the influence of different solid-liquid interface energies and the concentration of the melt on the arising patterns.

Using the same approach, four of the five theoretically predicted patterns from[176,177] are reported by Hötzer et al. in[178] in 3D simulations of ternary eutectics. In[73] this work is continued for the ternary eutectic system Al-Ag-Cu to examine patterns above and below the solubility shift of the system. Therefore, large-scale simulations with a domain size of 2420 × 2420 × 1474 and up to 84700 cores for 7 h are executed on high performance clusters. Steinmetz et al.[74] find a good quantitative accordance of these simulations[73] and further results with experimental micrographs. For comparison, principle component analysis (PCA) based on two-point correlations is applied. The PCA indicates the necessity of large-scale simulations to obtain statistical volume elements. Based on a Jackson-Hunt analysis in 3D for hexagonal structures, a systematic study is conducted in[85] to investigate the influence of the domain size on the pattern evolution. In large-scale simulations the formation of different aligned hexagonal regions is observed, which can not be resolved in smaller domains. To prove the experimental evidence of spiral growth from longitudinal sections in ternary eutectics reported by Genau et al.,[195] Hötzer et al.[170] discuss 3D large-scale phase-field simulations by exploiting the tilt instability from 2D. A helical growth of two intertwined phases embedded in a matrix phase is identified. Another approach to simulate ternary eutectics with the PFM is presented by Danilov and Nestler[214] with finite elements.

3.3 *Cell structures and eutectic colonies*

During directional solidification, cell structures can grow in systems with two or more components, if the concentration of the melt is different from the concentrations of the solidified phases. In this case, the melt enriches with the non-solidified components in the vicinity of the growth front.

Therefore, an impurity-driven Mullins-Sekerka instability[7–9,215] can occur,[216] leading to an enhanced growth of particular parts of the solidification front. An example of this dendrite-like behavior can be seen in.[217–219]

Phase-field studies of two-phase cell growth are reported by Losert et al.[68] Further studies with the PFM and stability analyses are conducted by Lan and co-workers,[220–222] who classified different shapes of the formations depending on the growth velocity and cell spacing. The influence of convection and gravity on cell growth is studied by Lan et al. in.[223]

For systems with more than two components, eutectic colonies can evolve, as combined mechanism of cell and eutectic growth for multiphase systems, as shown in experimental works of.[224,225] Raj and Locci[226] investigate the size of colonies depending on the growth rate experimentally.

A theory for the growth behavior of two-phase eutectic cells is proposed by Himemiya.[227] With this model, cellular-eutectic and dendritic-eutectic growth can be distinguished and a phase selection map along the eutectic groove can be derived by using models for two-phase and three-phase planar eutectic growth together.

Plapp and Karma describe the initial stages of eutectic colony formation by the instability of a thin lamellar eutectic interface in the presence of a ternary impurity with a linear instability analysis[228] in 1999. The behavior of the morphological instability wavelength and the critical onset velocity is analogous to those of the Mullins-Sekerka instability of a monophase planar interface,[7–9,215] which is experimentally shown for CBr_4-C_2Cl_6 by.[216] A phase-field simulation of the transition between lamellar eutectic growth and cell growth is reported by Boettinger et al.[229] In 2002, Plapp and Karma present an isotropic phase-field model for eutectic solidification in the presence of ternary impurities, using large-scale simulations in 2D to study the formation of colonies[230] and compare the results with the theory in.[228] A summary of the work in the field of eutectic colonies is provided in.[231] In,[232] the model of [230] is extended to consider anisotropy of the solid-solid and solid-liquid interfaces, and the effect of the growth fronts in multiphase alloys is studied. A definite cell spacing adjusts in the system, which is not observed in the isotropic systems.

A special arrangement of eutectic colonies are spiral dendrites. In this complex spatial arrangement, two-phase eutectics rotate around each other in a conical spiral. Akamatsu et al. experimentally report a real-time observation of this kind of growth for the ternary eutectic alloy SCN-DC-NA.[233] In 2014, Akamatsu et al.[234] compare their results with theoretical approaches of Ivantsov[121] and Jackson-Hunt.[10] The growth pattern of spiral dendrites is investigated simulatively with the PFM by Pusztai et al.[235] Besides the visual accordance between the experimental results and simulations, a scaling of the tip radius with the interface free energy and the kinetic anisotropy is found. The influence of the surface energy anisotropies on the formation of two-phase spiral dendrites is further examined by Ratkai et al. in.[236] They conclude that an observation of this type of formation without anisotropy is unlikely in contrast to the expectations of Akamatsu et al.[233,234]

4 Structure Analysis and Validation

To compare experimental, theoretical and simulative results, various methods have been established. In the following, a selection of different analytical and statistical validation methods in the context of the phase-field method are presented.

4.1 *Analytical validation methods*

With the analytical Zener relationship,[237] the isotropic growth of a nucleus in an infinite domain of an undercooled melt can be calculated. The stability of the solid-liquid interface can be described with the Mullins-Sekerka theory.[7–9] For isothermal dendritic growth, an analytical approach of the tip velocity is derivated by Ivantsov[121] in 1947. A continuing mathematical description is presented by Horvay and Cahn[238] and a general overview is given in.[239] The comparison between the PFM and this approach is exemplary, as shown in.[87,123] In the PFM, the interface between the phases is artificially enlarged. To ensure the correctness of this diffuse interface, the accordance with the sharp-interface solution is shown by several authors.[19–26,86] Choudhury et al.[240] present results of dendritic solidification obtained with phase-field as well as cellular automaton models.

Experiments indicate that the lamellar spacing of eutectics is determined by the growth conditions during the directional solidification like growth velocity and undercooling. Jackson and Hunt[10] describe the interaction between the growth velocity v, the undercooling ΔT and the spacing λ in lamellar

and rod eutectics. A relationship of the form $\Delta T = Av\lambda + B/\lambda$ is formulated with the constants A and B depending on the physical parameters of the considered system. Hunt et al.[149] and Langer[66] study the stability of lamellar eutectic growth and define a criterion for stable growth, if the lamellar spacing λ is larger than a critical value λ_{JH}. This critical spacing is coincident with the point of minimum undercooling ΔT of the solidification front. The minimum undercooling at λ_{JH} can be described as $\Delta T = 2\sqrt{ABv}$. McCartney et al.[150] extend the theory from binary to ternary two-phase growth. Dantzig and Rappaz give a theoretical overview of this topic in their book *Solidification*.[6]

Experimental investigations of the Jackson-Hunt theory for two-phase systems are conducted by[156,157,173,174,181–183,191,209,210,241–244] and a simulative phase-field study by.[20,79,165] Himemiya and Umeda[179] derive a Jackson-Hunt theory for a three-phase ternary eutectic in three dimensions for idealized patterns. An accordance between experiments of the ternary eutectic system Al-Ag-Cu and the eutectic Jackson-Hunt theory is mentioned by Böyük et al.[175]

Two-dimensional simulations based on the phase-field method are analyzed by Choudhury et al.,[172] whereas Steinmetz et al.[85] show a three-dimensional study for a hexagonal structure of a three-phase ternary eutectic. A theory for multiple components is introduced by Catalina et al.[245]

4.2 *Statistical validation methods*

To investigate the local arrangement of different phases, nearest neighbor statistics have been established. The application of this method for solidification processes is demonstrated in.[73,159,189,190,195,246] However, in[73] it is pointed out that this method can lead to ambiguous results. A shape factor can be used to classify the form of a rod, e.g. in micrographs of the ternary eutectic system Al-Ag-Cu by Dennstedt and Ratke.[196] To describe the spatial arrangement of different phases, pole plots[194,197] and principal component analysis based on two-point correlations[74,247,248] are used.

Other statistical methods may be interface lengths, triple point densities or phase fractions.

5 Related Aspects

In the previous sections, models and their applications, to solve the phase-field equations coupled with temperature and/or concentration for pure, binary and ternary systems are introduced. In this section, further related aspects to improve the understanding of the solidification process are presented. Afterwards, an overview of the ongoing progress to efficiently solve the PFM is given.

Most technically utilized alloys consist of multiple components. Impurities and small amounts of additional chemical elements, do affect the mechanical properties. First simulations of multicomponent systems of more than three components are displayed in[102] for the system Ni-Al-Co-Cr-Ti with 1D simulations and in[111] for Mg-Al-Mn-Zn with 2D simulations.

The influence of nucleation on the microstructure evolution during solidification is studied in.[32,33,57,65,111,205,249–254] To employ nucleation in the PFM, different approaches are used.[255] Nuclei can be set depending on given criterion, like the local temperature or concentration, or set randomized. Also random disturbances in the simulated fields can be imprinted or the diffuse interface can be exploited.

The influence of convection on the solidification is investigated for 2D in[223,256–261] and for 3D in.[262] In the work of Rojas et al.,[260] the growing dendrites are modeled as rigid bodies in a shear flow. Takaki et al.[261] provide an extension of this work by adding a gravity force.

The inclusion of further physical aspects to the models as well as the bridging of different length and timescales result in high computational effort. To handle this effort and compute the solidification process efficiently as well as on large domains, various optimization techniques are developed.

Optimizations on different levels, like the parameters, models, numerics, algorithms, parallelization and vectorization, are presented in the following. By exploiting the symmetry of a dendrite using Hill tetrahedrons, Berghoff et al.[263] show a reduction method to reduce the calculation cells by a factor of 96. Furthermore, an efficient way to simulate a single dendrite with the PFM by systematically rescaling the simulation domain, starting from a nucleus of a molecular dynamics (MD) or a phase-field crystal simulation (PFC) to a final dendrite is shown. This allows to bridge the scales between PFC with a nucleus of 50 Å in diameter and PFM with a final dendrite of 1 μm size.

Model optimizations to reduce the thickness of the compute intensive interface are introduced by Karma et al.[86,87] Kim et al.[88] exploit the limited number of nearest neighbors to reduce the number of locally required order parameters. With this, the required memory and the number of evolution equations is limited to a constant value.

Hötzer et al.[73] used an approach to exploit the differences in time scales of mass diffusion between solid and liquid and temperature diffusion. The findings allow to apply a moving window technique[128,230] as well as a frozen temperature approach.[229,264] To increase the numerical time step

Moving window technique: A technique to reduce the computational effort by systematically shifting the simulation domain, to focus on areas with the highest interest like e.g. the solidification front.

Frozen temperature approach: An approach to reduce the computational effort for solidification processes, in which the temperature diffusion is multiple times faster compared to the considered slower diffusion of the concentration. Therefore the effect of the temperature evolution can be neglected for the simulation of the solidification.

width, beside the commonly used explicit Euler scheme for the time discretion, implicit schemes are applied in[265–272] for two-dimensional and in[273] for three-dimensional dendritic growth. Semi-implicit approaches are presented by Vanherpe et al.[274] as well as Kornhuber and Krause.[275]

Adaptive mesh refinement (AMR) techniques, to locally enhance the resolution of the domain in local regions with larger gradients, are applied in[137,276,277] for dendritic growth, in[220,278] for cellular growth and in[70] for lamellar growth. An overview of different dynamic AMR algorithms is presented by Provatas et al. in.[34]

Vondrous et al.[279] and Bauer et al.[199] propose different optimizations of the used models and parameters to reduce the number of equations by classifying the current status of each cell in the domain. Both applied buffering techniques are capable of storing multiple required values, like gradients and staggered grid data. In 2008. Eiken[136,144] used various optimizations techniques to run large three dimensional simulations of dendritic growth on a single CPU.

To distribute the calculations and to solve the numerical methods parallel on multiple cores, commonly domain decomposition techniques are applied, with the message passing interface (MPI) to exchange the boundaries (ghost layers) between subdomains. Different approaches for the ghost layer exchange are shown in.[129,138,141,199,263,271,279,280] Accelerator cards like GPUs are employed in[138,141,280] for phase-field simulations to reduce the calculation time. A shared memory parallelization approach based on OpenMP is presented by Altenfeld et al.[281] A hybrid approach of the parallelization with MPI and OpenMP is presented in.[64,253,271]

In 1993, Wheeler et al.[91] achieved 200 MFLOPS on a single processor by vectorizing their code. Systematic node level performance engineering techniques are presented by Bauer et al.[199] to optimize the calculation of the phase-field evolution equations. With these techniques and an explicit vectorization, 25% peak performance on a single node of the SuperMUC supercomputer is achieved.

6 Conclusion and Perspectives

In the last decades, the phase-field method has been established for simulating solidification processes. Several aspects of solidification are modeled and studied. Single aspects can already be simulated in qualitative and quantitative accordance with theoretical and experimental results. However, the complex spatial interplay of different structures, particularly in 3D as well as the combination of various physical processes, is still challenging. These challenges have to be mastered to study the interaction for example, of:

- Solidified structures with external forces, like gravity, convection and mechanics
- Different solidified spatial structures, like the combined growth of eutectics, dendrites and eutectic colonies
- Multiple alloy elements (more than ternary)
- Solidification in combination with solid-solid phase transitions
- Physically correct nucleation models with pattern evolution

The investigations of such phenomena, are still limited due to the lack of suitable comprehensive models as well as the computational power of current supercomputers. The prediction of Warren and Boettinger[143] from 1995 that by using such methods "*... we must wait perhaps 10 y[ea] r[s] for computers to reach the power needed to do a large scale asymptotically, correct simulations ...*", is still not fulfilled today. Even if these calculations are possible today, the analysis of the results would require novel big data approaches and machine learning techniques. The cooperation of different scientific fields, following the idea of integrated computational materials engineering (ICME),[282] are in the line to address complex challenges more efficiently.

Ghost layers: Additional cells at the boundaries of a calculation block to enable the communication in the distributed parallel case between the blocks to calculate the gradients.

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References

1. C.T. Rios, S. Milenkovic, S. Gama and R. Caram, Influence of the growth rate on the microstructure of a Nb-Al-Ni ternary eutectic, *Journal of Crystal Growth* **237,** 90–94 (2002).
2. U. Böyük and N. Maraşlı, Dependency of eutectic spacings and microhardness on the temperature gradient for directionally solidified Sn-Ag-Cu lead-free solder, *Materials Chemistry and Physics* **119**(3), 442–448 (2010).
3. U. Böyük, Physical and mechanical properties of Al-Si-Ni eutectic alloy, *Metals and Materials International* **18**(6), 933–938 (2012).
4. E.C. Çadırlı, U. Böyük, H. Kaya and N. Maraşlı, Determination of mechanical, electrical and thermal properties of the Sn-Bi-Zn ternary alloy, *Journal of Non-Crystalline Solids* **357**(15), 2876–2881 (2011).
5. E. Çadırlı, U. Böyük, H. Kaya and N. Maraşlı, Effect of solidification parameters on the microstructure of directionally solidified Sn-Bi-Zn lead-free solder, *Metals and Materials International* **18**(2), 349–354 (2012).
6. J.A. Dantzig and M. Rappaz, *Solidification*, EPFL press (2009).

MFLOPS: Stands for mega floating point operations per second a measuring number to quantify the performance of a code.

7. W.W. Mullins and R.F. Sekerka, Morphological stability of a particle growing by diffusion or heat flow, *Journal of Applied Physics* **34**(2), 323–329 (1963).
8. W.W. Mullins and R.F. Sekerka, Stability of a planar interface during solidification of a dilute binary alloy, *Journal of Applied Physics* **35**(2), 444–451 (1964).
9. R.F. Sekerka, Morphological stability, *Journal of Crystal Growth* **3**, 71–81 (1968).
10. K.A. Jackson and J.D. Hunt, Lamellar and rod eutectic growth, *Transactions of the Metallurgical Society of AIME* **236**, 1129–1142 (1966).
11. K. Fisher and W. Kurz, Fundamentals of solidification, *Trans Tech Publications* (1986).
12. D.A. Kessler, J. Koplik and H. Levine, Pattern selection in fingered growth phenomena, *Advances in Physics* **37**(3), 255–339 (1988).
13. R.F. Sekerka, Role of instabilities in determination of the shapes of growing crystals, *Journal of Crystal Growth* **128**(1), 1–12 (1993).
14. W.J. Boettinger, S.R. Coriell, A.L. Greer, A. Karma, W. Kurz, M. Rappaz and R. Trivedi, Solidification microstructures: Recent developments, future directions, *Acta Materialia* **48**(1), 43–70 (2000).
15. S.H. Davis, *Theory of solidification*. Cambridge University Press, 2001.
16. R.F. Sekerka, Morphology: From sharp interface to phase field models, *Journal of Crystal Growth* **264**(4), 530–540 (2004).
17. G.E. Moore et al., Cramming more components onto integrated circuits, *Proceedings of the IEEE* **86**(1), 82–85 (1998).
18. N. Moelans, B. Blanpain and P. Wollants, An introduction to phase-field modeling of microstructure evolution, *Calphad* **32**(2), 268–294 (June 2008).
19. A.A. Wheeler, B.T. Murray and R.J. Schaefer, Computation of dendrites using a phase field model, *Physica D: Nonlinear Phenomena* **66**(1–2), 243–262 (1993).
20. A. Karma, Phase-field model of eutectic growth, *Physical Review E* **49**(3), 2245 (1994).
21. K.R. Elder, F. Drolet, J.M. Kosterlitz and M. Grant, Stochastic eutectic growth, *Physical Review Letters* **72**(5), 677 (1994).
22. J. Tiaden, B. Nestler, H. Diepers and I. Steinbach, The multiphase-field model with an integrated concept for modelling solute diffusion, *Physica D: Nonlinear Phenomena* **115**(1), 73–86 (1998).
23. H. Garcke, B. Nestler and B. Stoth, On anisotropic order parameter models for multi-phase systems and their sharp interface limits, *Physica D: Nonlinear Phenomena* **115**(1), 87–108 (1998).
24. A. Karma and W.J. Rappel, Phase-field model of dendritic sidebranching with thermal noise, *Physical Review E* **60**(4), 3614 (1999).
25. K.R. Elder, M. Grant, N. Provatas and J.M. Kosterlitz, Sharp interface limits of phase-field models, *Physical Review E* **64**(2), 021604 (2001).
26. K. Vetsigian and N. Goldenfeld, Computationally efficient phase-field models with interface kinetics, *Physical Review E* **68**(6), 060601 (2003).
27. J. Fan, M. Greenwood, M. Haataja and N. Provatas, Phase-field simulations of velocity selection in rapidly solidified binary alloys, *Physical Review E* **74**(3), 031602 (2006).
28. R. Qin and H. Bhadeshia, Phase field method, *Materials Science and Technology* **26**(7), 803–811 (2010).
29. W.J. Boettinger, J.A. Warren, C. Beckermann and A. Karma, Phase-field simulation of solidification 1, *Annual Review of Materials Research* **32**(1), 163–194 (2002).
30. L. Chen, Phase-field models for microstructure evolution, *Annual Review of Materials Research* **32**(1), 113–140 (2002).
31. K. Thornton, J. Ågren and P.W. Voorhees, Modelling the evolution of phase boundaries in solids at the meso- and nano-scales, *Acta Materialia* **51**(19), 5675–5710 (2003).
32. U. Hecht, L. Gránásy, T. Pusztai, B. Böttger, M. Apel, V. Witusiewicz, L. Ratke, J. De Wilde, L. Froyen, D. Camel, et al., Multiphase solidification in multicomponent alloys, *Materials Science and Engineering: R: Reports* **46**(1), 1–49 (2004).
33. L. Gránásy, T. Pusztai and J. Warren, Modelling polycrystalline solidification using phase field theory, *Journal of Physics: Condensed Matter* **16**(41), R1205 (2004).
34. N. Provatas, M. Greenwood, B. Athreya, N. Goldenfeld and J. Dantzig, Multiscale modeling of solidification: Phase-field methods to adaptive mesh refinement, *International Journal of Modern Physics B* **19**(31), 4525–4565 (2005).
35. L. Granasy, T. Pusztai, T. Börzsönyi, G. Toth, G. Tegze, J.A. Warren and J.F. Douglas, Phase field theory of crystal nucleation and polycrystalline growth: A review, *Journal of Materials Research* **21**(02), 309–319 (2006).
36. I. Singer-Loginova and H. Singer, The phase field technique for modeling multiphase materials, *Reports on Progress in Physics* **71**(10), 106501 (2008).
37. H. Emmerich, Advances of and by phase-field modelling in condensed-matter physics, *Advances in Physics* **57**(1), 1–87 (2008).
38. H. Emmerich, Phase-field modelling for metals and colloids and nucleation therein—an overview, *Journal of Physics: Condensed Matter* **21**(46), 464103 (2009).
39. M. Asta, C. Beckermann, A. Karma, W. Kurz, R. Napolitano, M. Plapp, G. Purdy, M. Rappaz and R. Trivedi, Solidification microstructures and solid-state parallels: Recent developments, future directions, *Acta Materialia* **57**(4), 941–971 (2009).
40. I. Steinbach, Phase-field models in materials science, *Modelling and Simulation in Materials Science and Engineering* **17**(7), 073001 (2009).
41. B. Nestler and A. Choudhury, Phase-field modeling of multi-component systems, *Current Opinion in Solid State and Materials Science* **15**(3), 93–105 (June 2011).

42. S. Akamatsu and M. Plapp, Eutectic and peritectic solidification patterns, *Current Opinion in Solid State and Materials Science* (2015).
43. N. Provatas and K. Elder, *Phase-field methods in materials science and engineering*, John Wiley & Sons (2011).
44. W. Kurz and P. Sahm, *Gerichtet erstarrte eutektische Werkstoffe: Herstellung, Eigenschaften und Anwendungen von In-situ-Verbundwerkstoffen* (Springer, 1975).
45. D.M. Herlach and R. Kirchheim, editors, *Phase Transformations in Multicomponent Melts*, Wiley-VCH Verlag GmbH & Co. KGaA, 1. auflage edition, 10 (2008).
46. J.S. Rowlinson, Translation of J.D. van der Waals' "the thermodynamik theory of capillarity under the hypothesis of a continuous variation of density", *Journal of Statistical Physics* **20**(2), 197–200 (1979).
47. On the theory of superconductivity, In D.T. Haar, editor, *Collected Papers of L.D. Landau*, Pergamon, pages 546–568 (1965).
48. L.D. Landau and I.M. Khalatikow, The selected works of l.d. landau (engl. transl.), (1963).
49. J.W. Cahn and J.E. Hilliard, Free energy of a nonuniform system. i. Interfacial free energy, *The Journal of Chemical Physics* **28**(2), 258–267 (1958).
50. J.W. Cahn, Free energy of a nonuniform system. ii. Thermodynamic basis, *The Journal of Chemical Physics* **30**(5), 1121–1124 (1959).
51. J.W. Cahn and J.E. Hilliard, Free energy of a nonuniform system, iii. nucleation in a two [U + 2010] component incompressible fluid, *The Journal of Chemical Physics* **31**(3), 688–699 (1959).
52. J.E. Hilliard and J.W. Cahn, On the nature of the interface between a solid metal and its melt, *Acta Metallurgica* **6**(12), 772–774 (1958).
53. S.M. Allen and J.W. Cahn, A microscopic theory for antiphase boundary motion and its application to antiphase domain coarsening, *Acta Metallurgica* **27**(6), 1085–1095 (1979).
54. G. Caginalp and P. Fife, Phase-field methods for interfacial boundaries, *Physical Review B* **33**(11), 7792 (1986).
55. G. Caginalp and P. Fife, Higher-order phase field models and detailed anisotropy, *Physical Review B* **34**(7), 4940 (1986).
56. A.A. Wheeler, W.J. Boettinger and G.B. McFadden, Phase-field model for isothermal phase transitions in binary alloys, *Phys. Rev. A* **45**, 7424–7439 (May 1992).
57. R. Kobayashi, Modeling and numerical simulations of dendritic crystal growth, *Physica D: Nonlinear Phenomena* **63**(3), 410–423 (1993).
58. B. Grossmann, K.R. Elder, M. Grant and J.M. Kosterlitz, Directional solidification in two and three dimensions, *Physical Review Letters* **71**(20), 3323 (1993).
59. G. Caginalp and W. Xie, Phase-field and sharp-interface alloy models, *Physical Review E* **48**(3), 1897 (1993).
60. A. A. Wheeler and G. B. McFadden, A ξ-vector formulation of anisotropic phase-field models: 3D asymptotics, *European Journal of Applied Mathematics* **7**(04), 367–381 (1996).
61. R. Folch and M. Plapp, Towards a quantitative phase-field model of two-phase solidification, *Physical Review E* **68**(1), 010602 (2003).
62. O. Penrose and P.C. Fife, Thermodynamically consistent models of phase-field type for the kinetic of phase transitions, *Physica D: Nonlinear Phenomena* **43**(1), 44–62 (1990).
63. S.L. Wang, R.F. Sekerka, A.A. Wheeler, B.T. Murray, S.R. Coriell, R.J. Braun, and G.B. McFadden, Thermodynamically-consistent phase-field models for solidification, *Physica D: Nonlinear Phenomena* **69**(1), 189–200 (1993).
64. B. Nestler, H. Garcke and B. Stinner, Multicomponent alloy solidification: Phase-field modeling and simulations, *Physical Review E* **71**, *041609* (2005).
65. A. Choudhury, B. Nestler, A. Telang, M. Selzer and F. Wendler, Growth morphologies in peritectic solidification of Fe-C: A phase-field study, *Acta Materialia* **58**(10), 3815–3823 (2010).
66. J.S. Langer, Eutectic solidification and marginal stability, *Phys. Rev. Lett.* **44**, 1023–1026 (Apr 1980).
67. S.G. Kim, W.T. Kim, T. Suzuki and M. Ode, Phase-field modeling of eutectic solidification, *Journal of Crystal Growth* **261**(1), 135–158 (2004).
68. W. Losert, D.A. Stillman, H.Z. Cummins, P. Kopczyński, W.J. Rappel and A. Karma, Selection of doublet cellular patterns in directional solidification through spatially periodic perturbations, *Physical Review E* **58**(6), 7492 (1998).
69. Mathis Plapp and Marcus Dejmek, Stability of hexagonal solidification patterns, *EPL (Europhysics Letters)* **65**(2), 276 (2004).
70. R. Folch and M. Plapp, Quantitative phase-field modeling of two-phase growth, *Physical Review E* **72**(1), 011602 (2005).
71. M. Plapp, Unified derivation of phase-field models for alloy solidification from a grand-potential functional, *Physical Review E* **84**(3), 031601 (2011).
72. A. Choudhury and B. Nestler, Grand-potential formulation for multicomponent phase transformations combined with thin-interface asymptotics of the double-obstacle potential, *Physical Review E* **85**(2), 021602 (2012).
73. J. Hötzer, M. Jainta, P. Steinmetz, B. Nestler, A. Dennstedt, A. Genau, M. Bauer, H. Köstler and U. Rüde, Large scale phase-field simulations of directional ternary eutectic solidification, *Acta Materialia* **93**(0), 194–204 (2015).
74. P. Steinmetz, Y.C. Yabansu, J. Hötzer, M. Jainta, B. Nestler and S.R. Kalidindi, Analytics for microstructure datasets produced by phase-field simulations, *Acta Materialia* **103**, 192–203 (2016).
75. H.W. Alt and I. Pawlow, A mathematical model of dynamics of non-isothermal phase separation, *Physica D: Nonlinear Phenomena* **59**(4), 389–416 (1992).

76. A.A. Wheeler, G.B. McFadden, and W.J. Boettinger, Phase-field model for solidification of a eutectic alloy, In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, volume **452**, 495–525, The Royal Society (1996).
77. Z. Bi and R.F. Sekerka, Phase-field model of solidification of a binary alloy, *Physica A: Statistical Mechanics and its Applications* **261**(1), 95–106 (1998).
78. I. Steinbach, F. Pezzolla, B. Nestler, M. Seeßelberg, R. Prieler, G.J. Schmitz and J.L.L. Rezende, A phase field concept for multiphase systems, *Physica D: Nonlinear Phenomena* **94**(3), 135–147 (1996).
79. B. Nestler and A. Wheeler, A multi-phase-field model of eutectic and peritectic alloys: Numerical simulation of growth structures, *Physica D: Nonlinear Phenomena* **138**(1), 114–133 (2000).
80. H. Garcke, B. Nestler and B. Stinner, A diffuse interface model for alloys with multiple components and phases, *Siam J. Appl. Math.*, **64**(3), 775–779 (2004).
81. B. Nestler, A 3D parallel simulator for crystal growth and solidification in complex alloy systems, *Journal of Crystal Growth* **275**(1), e273–e278 (2005).
82. P.R. Cha, D.H. Yeon and J.K. Yoon, Phase-field model for multicomponent alloy solidification, *Journal of Crystal Growth* **274**(1), 281–293 (2005).
83. J. Eiken, B. Böttger and I. Steinbach, Multiphase-field approach for multicomponent alloys with extrapolation scheme for numerical application, *Physical Review E* **73**(6), 066122 (2006).
84. G. Caginalp, Stefan and hele-shaw type models as asymptotic limits of the phase-field equations, *Physical Review A* **39**(11), 5887 (1989).
85. P. Steinmetz, J. Hötzer, M. Kellner, A. Dennstedt and B. Nestler, Large-scale phase-field simulations of ternary eutectic microstructure evolution, *Computational Materials Science (submitted)* (2016).
86. A. Karma and W. Rappel, Phase-field method for computationally efficient modeling of solidification with arbitrary interface kinetics, *Physical Review E* **53**(4), R3017 (1996).
87. A. Karma and W. Rappel, Quantitative phase-field modeling of dendritic growth in two and three dimensions, *Physical Review E* **57**(4), 4323 (1998).
88. S.G. Kim, D.I. Kim, W.T. Kim and Y.B. Park, Computer simulations of two-dimensional and three-dimensional ideal grain growth, *Phys. Rev. E* **74**, 061605 (Dec 2006).
89. I. Steinbach and F. Pezzolla, A generalized field method for multiphase transformations using interface fields, *Physica D: Nonlinear Phenomena* **134**(4), 385–393 (1999).
90. M. Selzer, *Mechanische und Strömungsmechanische Topologieoptimierung mit der Phasenfeldmethode*, PhD thesis, Karlsruhe, Karlsruher Institut für Technologie (KIT), Diss., 2014 (2014).
91. A.A. Wheeler, W.J. Boettinger and G.B. McFadden, Phase-field model of solute trapping during solidification, *Physical Review E* **47**(3), 1893 (1993).
92. W.J. Boettinger, A.A. Wheeler, B.T. Murray and G.B. McFadden, Prediction of solute trapping at high solidification rates using a diffuse interface phase-field theory of alloy solidification, *Materials Science and Engineering: A* **178**(1), 217–223 (1994).
93. N.A. Ahmad, A.A. Wheeler, W.J. Boettinger and G.B. McFadden, Solute trapping and solute drag in a phase-field model of rapid solidification, *Physical Review E* **58**(3), 3436 (1998).
94. M. Conti and M. Fermani, Interface dynamics and solute trapping in alloy solidification with density change, *Physical Review E* **67**(2), 026117 (2003).
95. H. Wang, F. Liu, G.J. Ehlen, and D.M. Herlach, Application of the maximal entropy production principle to rapid solidification: A multi-phase-field model, *Acta Materialia* **61**(7), 2617–2627 (2013).
96. A. Karma, Phase-field formulation for quantitative modeling of alloy solidification, *Physical Review Letters* **87**(11), 115701 (2001).
97. B. Echebarria, R. Folch, A. Karma and M. Plapp, Quantitative phase-field model of alloy solidification, *Physical Review E* **70**(6), 061604 (2004).
98. S.G. Kim, A phase-field model with antitrapping current for multicomponent alloys with arbitrary thermodynamic properties, *Acta Materialia* **55**(13), 4391–4399 (2007).
99. A. Carré, B. Böttger, and M. Apel, Implementation of an antitrapping current for a multicomponent multiphase-field ansatz, *Journal of Crystal Growth* **380**, 5–13 (2013).
100. P.R. Cha, D.H. Yeon and J.K. Yoon, A phase field model for isothermal solidification of multicomponent alloys, *Acta Materialia* **49**(16), 3295–3307 (2001).
101. U. Grafe, B. Böttger, J. Tiaden and S. Fries. Coupling of multicomponent thermodynamic databases to a phase field model: Application to solidification and solid state transformations of superalloys, *Scripta Materialia* **42**(12), 1179–1186 (2000).
102. U. Grafe, B. Böttger, J. Tiaden and S. Fries, Simulations of the initial transient during directional solidification of multicomponent alloys using the phase field method, *Modelling and Simulation in Materials Science and Engineering* **8**(6), 871–879 (2000).
103. H. Kobayashi, M. Ode, S.G. Kim, W.T. Kim and T. Suzuki, Phase-field model for solidification of ternary alloys coupled with thermodynamic database, *Scripta Materialia* **48**(6), 689–694 (2003).
104. R.S. Qin and E.R. Wallach, A phase-field model coupled with a thermodynamic database, *Acta Materialia*, **51**(20), 6199–6210 (2003).
105. R.S. Qin, E.R. Wallach and R.C. Thomson, A phase-field model for the solidification of multicomponent and multiphase alloys, *Journal of Crystal Growth* **279**(1), 163–169 (2005).
106. I. Steinbach, B. Böttger, J. Eiken, N. Warnken and S.G. Fries, Calphad and phase-field modeling: A successful liaison, *Journal of Phase Equilibria and Diffusion* **28**(1), 101–106 (2007).

107. S.Z. Zhang, R.J. Zhang, X.H. Qu, W. Fang and M.Z. Liu. Phase field simulation for non-isothermal solidification of multicomponent alloys coupled with thermodynamics database, *Transactions of Nonferrous Metals Society of China* **23**(8), 2361–2367 (2013).
108. G.J. Schmitz, B. Zhou, B. Böttger, S. Klima and J. Villain, Phase-field modeling and experimental observation of microstructures in solidifying Sn-Ag-Cu solders, *Journal of Electronic Materials* **42**(8), 2658–2666 (2013).
109. A. Choudhury, M. Kellner and B. Nestler, A method for coupling the phase-field model based on a grand-potential formalism to thermodynamic databases, *Current Opinion in Solid State and Materials Science* (0):– (2015).
110. B. Böttger, U. Grafe, D. Ma and S.G. Fries, Simulation of microsegregation and microstructural evolution in directionally solidified superalloys, *Materials Science and Technology* **16**(11–12), 1425–1428 (2000).
111. B. Böttger, J. Eiken and I. Steinbach, Phase field simulation of equiaxed solidification in technical alloys, *Acta Materialia* **54**(10), 2697–2704 (2006).
112. B. Böttger, J. Eiken, and M. Apel, Multi-ternary extrapolation scheme for efficient coupling of thermodynamic data to a multi-phase-field model, *Computational Materials Science,* **108**, Part B:283–292 (2015), Selected Articles from Phase-field Method 2014 International Seminar.
113. R.F. Almgren, Second-order phase field asymptotics for unequal conductivities, *SIAM Journal on Applied Mathematics* **59**(6), 2086–2107 (1999).
114. S. Akamatsu and M. Plapp. Eutectic and peritectic solidification patterns, *Current Opinion in Solid State and Materials Science* (2015).
115. J.W. Cahn. On spinodal decomposition, *Acta Metallurgica* **9**(9), 795–801 (1961).
116. J.S. Langer, Theory of spinodal decomposition in alloys, *Annals of Physics* **65**(1), 53–86 (1971).
117. R.W. Balluffi, S. Allen and W. Carter, *Kinetics of Materials*, John Wiley & Sons (2005).
118. M. Tang, A. Karma, et al., Surface modes of coherent spinodal decomposition, *Physical Review Letters* **108**(26), 265701 (2012).
119. F. Wang, A. Choudhury, C. Strassacker and B. Nestler, Spinodal decomposition and droplets entrapment in monotectic solidification, *The Journal of Chemical Physics* **137**(3), 034702 (2012).
120. F. Wang, A. Choudhury, M. Selzer, R. Mukherjee and B. Nestler, Effect of solutal marangoni convection on motion, coarsening, and coalescence of droplets in a monotectic system, *Physical Review E* **86**(6), 066318 (2012).
121. G.P. Ivantsov, The temperature field around a spherical, cylindrical, or pointed crystal growing in a cooling solution, In *Dokl. Akad. Nauk SSSR* volume **58**, pages 567–569 (1947).
122. S.L. Wang and R.F. Sekerka, Computation of the dendritic operating state at large supercoolings by the phase field model, *Phys. Rev. E* **53**, 3760–3776 (Apr 1996).
123. A. Karma and W. Rappel, Numerical simulation of three-dimensional dendritic growth, *Physical Review Letters* **77**(19), 4050 (1996).
124. W.J. Boettinger and J.A. Warren, The phase-field method: Simulation of alloy dendritic solidification during recalescence, *Metallurgical and Materials Transactions A* **27**(3), 657–669 (1996).
125. D. Ma and P.R. Sahm, Primary spacing in directional solidification, *Metallurgical and Materials Transactions A* **29**(13), 1113–1119 (1998).
126. N. Provatas, N. Goldenfeld and J. Dantzig. Efficient computation of dendritic microstructures using adaptive mesh refinement, *Physical Review Letters* **80**(15), 3308 (1998).
127. Y.T. Kim, N. Provatas, N. Goldenfeld, and J. Dantzig, Universal dynamics of phase-field models for dendritic growth, *Physical Review E* **59**(3), R2546 (1999).
128. H.J. Diepers, D. Ma and I. Steinbach, History effects during the selection of primary dendrite spacing, comparison of phase-field simulations with experimental observations, *Journal of Crystal Growth* **237–239**, Part 1: 149–153, (2002), The thirteenth international conference on Crystal Growth in conjunction with the eleventh international conference on Vapor Growth and Epitaxy.
129. W.L. George and J.A. Warren, A parallel 3D dendritic growth simulator using the phase-field method, *Journal of Computational Physics* **177**(2), 264–283 (2002).
130. L. Gránásy, T. Börzsönyi and T. Pusztai, Crystal nucleation and growth in binary phase-field theory, *Journal of Crystal Growth* **237**, 1813–1817 (2002).
131. L. Gránásy, T. Börzsönyi and T. Pusztai, Nucleation and bulk crystallization in binary phase field theory, *Physical Review Letters* **88**(20), 206105 (2002).
132. L. Gránásy, T. Pusztai, T. Börzsönyi, J. Warren and J. Douglas, A general mechanism of polycrystalline growth, *Nature Materials* **3**(9), 645–650 (2004).
133. D. Danilov and B. Nestler, Dendritic to globular morphology transition in ternary alloy solidification, *Physical Review Letters* **93**(21), 215501 (2004).
134. B. Böttger, J. Eiken, M. Ohno, G. Klaus, M. Fehlbier, R. Schmid-Fetzer, I. Steinbach and A. Bührig-Polaczek, Controlling microstructure in magnesium alloys: A combined thermodynamic, experimental and simulation approach, *Advanced Engineering Materials* **8**(4), 241–247 (2006).
135. P.K. Galenko, S. Reutzel, D.M. Herlach, S.G. Fries, I. Steinbach and M. Apel, Dendritic solidification in undercooled Ni-Zr-Al melts: Experiments and modeling, *Acta Materialia* **57**(20), 6166–6175 (2009).
136. J. Eiken, *A phase-field model for technical alloy solidification*, Shaker Verlag (2010).
137. N. Ofori-Opoku and N. Provatas, A quantitative multi-phase field model of polycrystalline alloy solidification, *Acta Materialia* **58**(6), 2155–2164 (2010).
138. T. Shimokawabe, T. Aoki, T. Takaki, A. Yamanaka, A. Nukada, T. Endo, Maruyama and S. Matsuoka,

Peta-scale phase-field simulation for dendritic solidification on the TSUBAME 2.0 supercomputer, *2011 International Conference for, High Performance Computing, Networking, Storage and Analysis (SC)*, pages 1–11 (2011).

139. A. Yamanaka, T. Aoki, S. Ogawa and T. Takaki, GPU-accelerated phase-field simulation of dendritic solidification in a binary alloy, *Journal of Crystal Growth* **318**(1), 40–45 (2011).
140. E. Wesner, A. Choudhury, A. August, M. Berghoff and B. Nestler, A phase-field study of large-scale dendrite fragmentation in Al-Cu, *Journal of Crystal Growth* **359**, 107–121 (2012).
141. T. Takaki, T. Shimokawabe, M. Ohno, A. Yamanaka and T. Aoki, Unexpected selection of growing dendrites by very-large-scale phase-field simulation, *Journal of Crystal Growth* **382**, 21–25 (2013).
142. T. Takaki, Phase-field modeling and simulations of dendrite growth, *ISIJ International* **54**(2), 437–444 (2014).
143. J.A. Warren and W.J. Boettinger, Prediction of dendritic growth and microsegregation patterns in a binary alloy using the phase-field method, *Acta Metallurgica et Materialia* **43**(2), 689–703 (1995).
144. J. Eiken, Private Communication (2015).
145. Tomohiro Takaki, Munekazu Ohno, Yasushi Shibuta, Shinji Sakane, Takashi Shimokawabe, and Takayuki Aoki. Two-dimensional phase-field study of competitive grain growth during directional solidification of polycrystalline binary alloy, *Journal of Crystal Growth*, page – (2016).
146. I. Loginova, G. Amberg and J. Ågren, Phase-field simulations of non-isothermal binary alloy solidification, *Acta Materialia* **49**(4), 573–581 (2001).
147. H. Emmerich, M. Jurgk and R. Siquieri, Multiscale simulations of microstructure selection in binary alloy solidification, *Physica Status Solidi (b)* **241**(9), 2128–2135 (2004).
148. H. Fredriksson and U. Akerlind, *Solidification and crystallization processing in metals and alloys*, John Wiley & Sons (2012).
149. J. Hunt, D. Hurle, K. Jackson and E. Jakeman, On the theory of the stability of lamellar eutectics, *Metallurgical and Materials Transactions B* **1**(1), 318–320 (1970).
150. D.G. McCartney, J.D. Hunt and R.M. Jordan, The structures expected in a simple ternary eutectic system: Part 1. theory, *Metallurgical Transactions A* **11**(8), 1243–1249 (1980).
151. S. R. Coriell, G.B. McFadden, P.W. Voorhees and R.F. Sekerka, Stability of a planar interface during solidification of a multicomponent system, *Journal of Crystal Growth* **82**(3), 295–302 (1987).
152. K. Kassner, A. Valance, C. Misbah and D. Temkin, New broken-parity state and a transition to anomalous lamellae in eutectic growth, *Physical Review E* **48**(2), 1091 (1993).
153. K. Kassner, C. Misbah and R. Baumann, Eutectic dynamics: A host of new states, *Physical Review E* **51**(4), R2751 (1995).
154. A. Karma and A. Sarkissian, Morphological instabilities of lamellar eutectics, *Metallurgical and Materials Transactions A* **27**(3), 635–656 (1996).
155. C. Caroli and G. Faivre, Lamellar eutectic growth: A discussion of the question of pattern selection, In *Solid State Phenomena*, volume **42**, pages 1–10, Trans Tech Publ, (1995).
156. G. Faivre, Morphological instabilities of lamellar eutectic growth fronts: A survey of recent experimental and numerical results, *Journal of Crystal Growth* **166**(1), 29–39 (1996).
157. M. Ginibre, S. Akamatsu and G. Faivre, Experimental determination of the stability diagram of a lamellar eutectic growth front, *Physical Review E* **56**, 780–796 (Jul 1997).
158. S. Akamatsu, S. Bottin-Rousseau and G. Faivre, Experimental evidence for a zigzag bifurcation in bulk lamellar eutectic growth, *Physical Review Letters* **93**(17), 175701 (2004).
159. S. Bottin-Rousseau, M. Perrut, C. Picard, S. Akamatsu and G. Faivre, An experimental method for the in situ observation of eutectic growth patterns in bulk samples of transparent alloys, *Journal of Crystal Growth* **306**(2), 465–472 (2007).
160. S. Akamatsu, S. Bottin-Rousseau, M. Şerefoğlu, and G. Faivre, Lamellar eutectic growth with anisotropic interphase boundaries: Experimental study using the rotating directional solidification method, *Acta Materialia* **60**(6–7), 3206–3214 (2012).
161. S. Akamatsu, S. Bottin-Rousseau, M. Şerefoğlu, and G. Faivre, A theory of thin lamellar eutectic growth with anisotropic interphase boundaries, *Acta Materialia* **60**(6–7), 3199–3205 (2012).
162. S. Akamatsu, S. Bottin-Rousseau, G. Faivre, S. Ghosh and M. Plapp, Lamellar eutectic growth with anisotropic interphase boundaries, *IOP Conference Series: Materials Science and Engineering* **84**(1), 012083 (2015).
163. Silvére Akamatsu and Henri Nguyen-Thi, In situ observation of solidification patterns in diffusive conditions, *Acta Materialia*, pages – (2016).
164. S. Rex, B. Böttger, V. Witusiewicz, and U. Hecht, Transient eutectic solidification in In-Bi-Sn: Two-dimensional experiments and numerical simulation, *Materials Science and Engineering: A* **413–414**, 249–254 (2005), International Conference on Advances in Solidification Processes.
165. M. Apel, B. Boettger, H.J. Diepers and I. Steinbach, 2D and 3D phase-field simulations of lamella and fibrous eutectic growth. *Journal of Crystal Growth*, 237–239, Part **1**, 154–158, (2002), The thirteenth international conference on Crystal Growth in conjunction with the eleventh international conference on Vapor Growth and Epitaxy.
166. S. Ghosh, A. Choudhury, M. Plapp, S. Bottin-Rousseau, G. Faivre and S. Akamatsu, Interphase anisotropy effects on lamellar eutectics: A numerical study, *Physical Review E* **91**, 022407 (Feb 2015).

167. S. Ghosh and M. Plapp, Influence of interphase anisotropy on lamellar eutectic growth patterns, *Transactions of the Indian Institute of Metals* **68**(6), 1235–1238 (2015).
168. S. Ghosh, *Effects of solid-solid boundary anisotropy on directional solidification microstructures*, PhD thesis, Ecole Polytechnqiue (2015).
169. M. Apel, B. Böttger, V. Witusiewicz, U. Hecht and I. Steinbach, Lamellar pattern formation during 2D-directional solidification of ternary eutectic alloys, *Solidification and Crystallization,* pages 271–279 (2004).
170. J. Hötzer, P. Steinmetz, M. Jainta, S. Schulz, M. Kellner, B. Nestler, A. Genau, A. Dennstedt, M. Bauer, H. Köstler and U. Rüde. Phase-field simulations of spiral growth during directional ternary eutectic solidification, *Acta Materialia* **106**, 249–259 (2016).
171. A. Choudhury, M. Geeta and B. Nestler, Influence of solid-solid interface anisotropy on three-phase eutectic growth during directional solidification, *EPL (Europhysics Letters)* **101**(2), 26001 (2013).
172. A. Choudhury, M. Plapp and B. Nestler, Theoretical and numerical study of lamellar eutectic three-phase growth in ternary alloys, *Physical Review E* **83**(5), 051608 (2011).
173. P.R. Sahm and M. Lorenz. Strongly coupled growth in faceted-nonfaceted eutectics of the monovariant type, *Journal of Materials Science* **7**(7), 793–806 (1972).
174. M.D. Rinaldi, R.M. Sharp and M.C. Flemings, Growth of ternary composites from the melt: Part II. *Metallurgical Transactions* **3**(12), 3139–3148 (1972).
175. U. Böyük, N. Maraşlı, H. Kaya, E. Çadırlı, and K. Keşlioğlu, Directional solidification of Al-Cu-Ag alloy, *Applied Physics A* **95**(3), 923–932 (2009).
176. M. Ruggiero and J. Rutter, Origin of microstructure in the 332 K eutectic of the Bi-In-Sn system, *Materials Science and Technology* **13**(1), 5–11 (1997).
177. D. Lewis, S. Allen, M. Notis and A. Scotch, Determination of the eutectic structure in the Ag-Cu-Sn system, *Journal of Electronic Materials* **31**(2), 161–167 (2002).
178. J. Hötzer, M. Jainta, P. Steinmetz, A. Dennstedt and B. Nestler, Die Vielfalt der Musterbildung in Metallen (2015).
179. T. Himemiya and T. Umeda, Three-phase planar eutectic growth models for a ternary eutectic system, *Materials Transactions, JIM* **40**(7), 665–674 (1999).
180. D.J.S. Cooksey and A. Hellawell, The Microstructures of Ternary Eutectic Alloys in the Systems Cd-Sn-(Pb, In, Tl), Al-Cu-(Mg, Zn, Ag) and Zn-Sn-Pb. *J. Inst. Met.* **95**, 183–187 (1967).
181. P.J. Fehrenbach, H.W. Kerr and P. Niessen, Unidirectional solidification of monovariant eutectic Cu-Mg-Ni alloys: I. Planar interface stability criterion, *Journal of Crystal Growth* **16**(3), 209–214 (1972).
182. P.J. Fehrenbach, H.W. Kerr and P. Niessen, Unidirectional solidification of monovariant Cu-Ni-Mg eutectic alloys II. Microstructures and Properties, *Journal of Crystal Growth* **18**(2), 151–158 (1973).
183. J.D. Holder and B.F. Oliver, The directional solidification of Pb-Sn-Cd alloys, *Metallurgical Transactions* **5**(11), 2423–2437 (1974).
184. M. Ruggiero and J. Rutter, Microstructure of Bi-Cd-Sn ternary eutectic, *Materials Science and Technology* **8**(6), 523–530 (1992).
185. M. Ruggiero and J. Rutter, Origin of microstructure in 350 K eutectic of Bi-In-Sn ternary system, *Materials Science and Technology* **11**(2), 136–142 (1995).
186. M. Ruggiero and J. Rutter, Microstructure of Bi-Cd-Pb ternary eutectic, *Materials Science and Technology* **12**(1), 7–11 (1996).
187. M. Ruggiero and J. Rutter, Formation of microstructure in the Cd-In-Sn ternary eutectic, *Materials Science and Technology* **14**(3), 177–181 (1998).
188. M. Ruggiero and J. Rutter, Formation of microstructure during solidification of Bi-Pb-Sn ternary eutectic, *Materials Science and Technology* **15**(10), 1110–1114 (1999).
189. M. Perrut, S. Akamatsu, S. Bottin-Rousseau and G. Faivre, Long-time dynamics of the directional solidification of rodlike eutectics, *Physical Review E* **79**(3), 032602 (2009).
190. M. Perrut, S. Bottin-Rousseau, G. Faivre and S. Akamatsu, Dynamic instabilities of rod-like eutectic growth patterns: A real-time study, *Acta Materialia* **61**(18), 6802–6808 (2013).
191. J. De Wilde, L. Froyen and S. Rex, Coupled two-phase [α (al) + θ (Al 2 Cu)] planar growth and destabilisation along the univariant eutectic reaction in Al-Cu-Ag alloys, *Scripta Materialia* **51**(6), 533–538 (2004).
192. D.G. Mccartney, R.M. Jordan and J.D. Hunt, The structures expected in a simple ternary eutectic system: Part II. the Al-Ag-Cu ternary system, *Metallurgical Transactions A* **11**(8), 1251–1257 (1980).
193. I. Sargin, *Invariant and univariant eutectic solidification in ternary alloys*, PhD thesis, Iowa State University (2015).
194. A.L. Genau and L. Ratke, Crystal orientation and morphology in Al-Ag-Cu ternary eutectic, *IOP Conference Series: Materials Science and Engineering* **27**(1), 012032 (2012).
195. A. Genau and L. Ratke, Morphological characterization of the Al-Ag-Cu ternary eutectic, *International Journal of Materials Research* **103**(4), 469–475 (April 2012).
196. A. Dennstedt and L. Ratke, Microstructures of directionally solidified Al-Ag-Cu ternary eutectics, *Transactions of the Indian Institute of Metals* **65**(6), 777–782 (2012).
197. A. Dennstedt, L. Ratke, A. Choudhury and B. Nestler, New metallographic method for estimation of ordering and lattice parameter in ternary eutectic systems, *Metallography, Microstructure, and Analysis* **2**(3), 140–147 (2013).
198. A. Dennstedt, L. Helfen, P. Steinmetz, B. Nestler and L. Ratke, 3d synchrotron imaging of a directionally solidified ternary eutectic, *Metallurgical and Materials Transactions A*, pages 1–4 (2015).

199. M. Bauer, J. Hötzer, P. Steinmetz, M. Jainta, M. Berghoff, F. Schornbaum, C. Godenschwager, H. Köstler, B. Nestler and U. Rüde, Massively parallel phase-field simulations for ternary eutectic directional solidification, *arXiv preprint arXiv: 1506.01684, accepted at SuperComputing 2015* (2015).
200. U. Böyük, S. Engin, and N. Maraşlı. Microstructural characterization of unidirectional solidified eutectic Al-Si-Ni alloy, *Materials Characterization* **62**(9), 844–851 (2011).
201. S. Engin, U. Böyük and N. Maraşlı, Determination of solid-liquid interfacial energy for a solid sn in equilibrium with a Sn-Ag-Zn eutectic liquid, *Current Applied Physics* **11**(4), 1060–1066 (2011).
202. F.S. Drolet, K.R. Elder, M. Grant and J.M. Kosterlitz, Phase-field modeling of eutectic growth, *Physical Review E* **61**(6), 6705 (2000).
203. B. Nestler and A. Wheeler, Phase-field modeling of multi-phase solidification, *Computer Physics Communications* **147**(1), 230–233 (2002).
204. B. Nestler, A. Wheeler and H. Garcke, Modelling of microstructure information and interface dynamics, *Computational Materials Science* **26**, 111–119 (2003).
205. J.R. Green, P.K. Jimack and A.M. Mullis, Phase field analysis of eutectic breakdown, *Metallurgical and Materials Transactions A* **38**(7), 1426–1432 (2007).
206. J. Kundin, E. Pogorelov and H. Emmerich, Phase-field modeling of the microstructure evolution and heterogeneous nucleation in solidifying ternary Al-Cu-Ni alloys, *Acta Materialia* **83**, 448–459 (2015).
207. D. Lewis, J. Warren, W. Boettinger, T. Pusztai and L. Gránásy, Phase-field models for eutectic solidification, *JOM* **56**(4), 34–39 (2004).
208. A. Parisi, M. Plapp, S. Akamatsu, S. Bottin-Rousseau, M. Perrut and G. Faivre, Three-dimensional phase-field simulations of eutectic solidification and comparison to in situ experimental observations, *Morphological stability of lamellar and rod eutectic growth, TMS (The Minerals, Metals & Materials Society)* (2005).
209. A. Parisi and M. Plapp, Stability of lamellar eutectic growth, *Acta Materialia* **56**(6), 1348–1357 (2008).
210. A. Parisi and M. Plapp, Defects and multistability in eutectic solidification patterns, *EPL* **90**(2), 26010 (2010).
211. M. Plapp, Three-dimensional phase-field simulations of directional solidification, *Journal of Crystal Growth* **303**(1), 49–57 (2007).
212. M. Perrut, A. Parisi, S. Akamatsu, S. Bottin-Rousseau, G. Faivre and M. Plapp, Role of transverse temperature gradients in the generation of lamellar eutectic solidification patterns, *Acta Materialia* **58**(5), 1761–1769 (2010).
213. A. Choudhury, Pattern-formation during self-organization in three-phase eutectic solidification, *Transactions of the Indian Institute of Metals*, pages 1–7 (2015).
214. D. Danilov and B. Nestler, Phase-field simulations of solidification in binary and ternary systems using a finite element method, *Journal of Crystal Growth* **275**(1), e177–e182 (2005).
215. R.F. Sekerka, A stability function for explicit evaluation of the mullins-sekerka interface stability criterion, *Journal of Applied Physics* **36**(1), 264–268 (1965).
216. S. Akamatsu and G. Faivre, Traveling waves, two-phase fingers, and eutectic colonies in thin-sample directional solidification of a ternary eutectic alloy, *Physical Review E* **61**(4), 3757 (2000).
217. S. Akamatsu and T. Ihle, Similarity law for the tilt angle of dendrites in directional solidification of non-axially-oriented crystals, *Physical Review E* **56**(4), 4479 (1997).
218. S. Akamatsu and G. Faivre, Anisotropy-driven dynamics of cellular fronts in directional solidification in thin samples, *Physical Review E* **58**(3), 3302 (1998).
219. S. Akamatsu, G. Faivre and S. Moulinet, The formation of lamellar-eutectic grains in thin samples, *Metallurgical and Materials Transactions A* **32**(8), 2039–2048 (2001).
220. C.W. Lan and Y.C. Chang, Efficient adaptive phase field simulation of directional solidification of a binary alloy, *Journal of Crystal Growth* **250**(3), 525–537 (2003).
221. C.W. Lan, C.J. Shih and W.T. Hsu, Long-time scale morphological dynamics near the onset of instability during directional solidification of an alloy, *Journal of Crystal Growth* **264**(1), 379–384 (2004).
222. C.W. Lan, C.J. Shih and M.H. Lee, Quantitative phase field simulation of deep cells in directional solidification of an alloy, *Acta Materialia* **53**(8), 2285–2294 (2005).
223. C.W. Lan, M.H. Lee, M.H. Chuang and C.J. Shih, Phase field modeling of convective and morphological instability during directional solidification of an alloy, *Journal of Crystal Growth* **295**(2), 202–208 (2006).
224. M. Durand-Charre and F. Durand, Effects of growth rate on the morphology of monovariant eutectics: MnSb-(Sb, Bi) and MnSb-(Sb, Sn), *Journal of Crystal Growth* **13**, 747–750 (1972).
225. U. Hecht, V. Witusiewicz and A. Drevermann, Coupled growth of A-l-Al2Cu eutectics in Al-Cu-Ag alloys. *IOP Conference Series: Materials Science and Engineering* **27**(1), 012029 (2012).
226. S.V. Raj and I.E. Locci, Microstructural characterization of a directionally-solidified ni-33 (at.%) Al-31Cr-3Mo eutectic alloy as a function of withdrawal rate, *Intermetallics* **9**(3), 217–227 (2001).
227. T. Himemiya, Growth models of two-phase eutectic cell in a ternary eutectic system: a phase selection map, *Materials Transactions, JIM* **40**(7), 675–684 (1999).
228. M. Plapp and A. Karma, Eutectic colony formation: A stability analysis, *Physical Review E* **60**(6), 6865 (1999).
229. W.J. Boettinger and J.A. Warren, Simulation of the cell to plane front transition during directional solidification at high velocity, *Journal of Crystal Growth* **200**(3), 583–591 (1999).
230. M. Plapp and A. Karma, Eutectic colony formation: A phase-field study, *Physical Review E* **66**(6), 061608 (2002).

231. A. Karma and M. Plapp, New insights into the morphological stability of eutectic and peritectic coupled growth, *JOM* **56**(4), 28–32 (2004).
232. A. Lahiri and A. Choudhury. Effect of surface energy anisotropy on the stability of growth fronts in multiphase alloys, *Transactions of the Indian Institute of Metals* **68**(6), 1053–1057 (2015).
233. S. Akamatsu, M. Perrut, S. Bottin-Rousseau and G. Faivre, Spiral two-phase dendrites, *Physical Review Letters* **104**, 056101 (Feb 2010).
234. S. Akamatsu, S. Bottin-Rousseau, G. Faivre and E.A. Brener, Scaling theory of two-phase dendritic growth in undercooled ternary melts, *Physical Review Letters* **112**(10), 105502 (2014).
235. T. Pusztai, L. Rátkai, A. Szállás and L. Gránásy, Spiraling eutectic dendrites, *Physical Review E* **87**(3), 032401 (2013).
236. L. Rátkai, A. Szállás, T. Pusztai, T. Mohri and L. Gránásy, Ternary eutectic dendrites: Pattern formation and scaling properties, *The Journal of Chemical Physics* **142**(15), 154501 (2015).
237. C. Zener, Theory of growth of spherical precipitates from solid solution, *Journal of Applied Physics* **20**(10), 950–953 (1949).
238. G. Horvay and J.W. Cahn, Dendritic and spheroidal growth, *Acta Metallurgica* **9**(7), 695–705 (1961).
239. S. McFadden and D.J. Browne, A generalised version of an ivantsov-based dendrite growth model incorporating a facility for solute measurement ahead of the tip, *Computational Materials Science* **55**, 245–254 (2012).
240. A. Choudhury, K. Reuther, E. Wesner, A. August, B. Nestler and M. Rettenmayr, Comparison of phase-field and cellular automaton models for dendritic solidification in Al-Cu alloy, *Computational Materials Science* **55**, 263–268, 2012.
241. S. Akamatsu, M. Plapp, G. Faivre and A. Karma. Pattern stability and trijunction motion in eutectic solidification, *Physical Review E* **66**(3), 030501 (2002).
242. S. Akamatsu, G. Faivre, M. Plapp and A. Karma, Overstability of lamellar eutectic growth below the minimum-undercooling spacing, *Metallurgical and Materials Transactions A* **35**(6), 1815–1828 (2004).
243. J. De Wilde, L. Froyen, V.T. Witusiewicz and U. Hecht, Two-phase planar and regular lamellar coupled growth along the univariant eutectic reaction in ternary alloys: An analytical approach and application to the Al-Cu-Ag system, *Journal of Applied Physics* **97**(11):, (2005).
244. S. Akamatsu, S. Bottin-Rousseau and G. Faivre, Determination of the jackson-hunt constants of the In-In2Bi eutectic alloy based on in situ observation of its solidification dynamics, *Acta Materialia* **59**(20), 7586–7591 (2011).
245. A.V. Catalina, P.W. Voorhees, R.K. Huff and A.L. Genau, A model for eutectic growth in multicomponent alloys, *IOP Conference Series: Materials Science and Engineering* **84**(1), 012085 (2015).
246. A. Dennstedt, A. Choudhury, L. Ratke and B. Nestler, Microstructures in a ternary eutectic alloy: Devising metrics based on neighbourhood relationships, *IOP Conference Series: Materials Science and Engineering*, accepted (2014).
247. S.R. Kalidindi, *Hierarchical Materials Informatics: Novel Analytics for Materials Data*, Butterworth-Heinemann, 1 edition, 8 (2015).
248. A. Choudhury, Private Communication (2015).
249. J.P Simmons, C. Shen and Y. Wang. Phase field modeling of simultaneous nucleation and growth by explicitly incorporating nucleation events, *Scripta Materialia* **43**(10), 935–942 (2000).
250. L. Gránásy and D. Oxtoby, Cahn-hilliard theory with triple-parabolic free energy, I. nucleation and growth of a stable crystalline phase, *The Journal of Chemical Physics* **112**(5), 2399–2409 (2000).
251. L. Gránásy and D. Oxtoby, Cahn-hilliard theory with triple-parabolic free energy, II. nucleation and growth in the presence of a metastable crystalline phase, *The Journal of Chemical Physics* **112**(5), 2410–2419 (2000).
252. R. Siquieri and H. Emmerich, Phase-field investigation of the nucleation kinetics and of the influence of convection on growth in peritectic systems, *Philosophical Magazine Letters* **87**(11), 829–837 (2007).
253. T. Pusztai, G. Tegze, G.I. Tóth, L. Környei, G. Bansel, Z. Fan and L. Gránásy, Phase-field approach to polycrystalline solidification including heterogeneous and homogeneous nucleation, *Journal of Physics: Condensed Matter* **20**(40), 404205 (2008).
254. J.A. Warren, T. Pusztai, L. Környei and L. Gránásy, Phase field approach to heterogeneous crystal nucleation in alloys, *Phys. Rev. B* **79**, 014204 (Jan 2009).
255. T.W. Heo and L.Q. Chen, Phase-field modeling of nucleation in solid-state phase transformations, *JOM* **66**(8), 1520–1528 (2014).
256. C. Beckermann, H.J. Diepers, I. Steinbach, A. Karma and X. Tong, Modeling melt convection in phase-field simulations of solidification, *Journal of Computational Physics* **154**(2), 468–496 (1999).
257. B. Nestler, A.A. Wheeler, L. Ratke and C. Stöcker, Phase-field model for solidification of a monotectic alloy with convection, *Physica D: Nonlinear Phenomena* **141**(1–2), 133–154 (2000).
258. G. Tegze, T. Pusztai and L. Gránásy, Phase field simulation of liquid phase separation with fluid flow, *Materials Science and Engineering: A* **413**, 418–422 (2005).
259. Z. Guo, J. Mi and P.S. Grant, Phase field simulation of multi-dendrite growth in a coupled thermal-solute-convective environment, In *IOP Conference Series: Materials Science and Engineering*, volume 33, page 012101, IOP Publishing (2012).
260. R. Rojas, T. Takaki, and M. Ohno, A phase-field-lattice boltzmann method for modeling motion and growth of a dendrite for binary alloy solidification in the presence of melt convection, *Journal of Computational Physics* **298**, 29–40 (2015).

261. T. Takaki, R. Rojas, M. Ohno, T. Shimokawabe and T. Aoki, GPU phase-field lattice boltzmann simulations of growth and motion of a binary alloy dendrite, *IOP Conference Series: Materials Science and Engineering* **84**(1), 012066 (2015).
262. Y. Lu, C. Beckermann and J.C. Ramirez, Three-dimensional phase-field simulations of the effect of convection on free dendritic growth, *Journal of Crystal Growth* **280**(1–2), 320–334 (2005).
263. M. Berghoff, M. Selzer, A. Choudhury, and B. Nestler, Efficient techniques for bridging from atomic to mesoscopic scale in phase-field simulations, *Journal of Computational Methods in Sciences and Engineering* **13**(5–6), 441–454 (2013).
264. Z. Bi and R.F. Sekerka, Phase field modeling of shallow cells during directional solidification of a binary alloy, *Journal of Crystal Growth* **237**, 138–143 (2002).
265. J. Rosam, *A fully implicit, fully adaptive multigrid method for multiscale phase-field modelling*, PhD thesis, Citeseer (2007).
266. J. Rosam, P.K. Jimack and A. Mullis, A fully implicit, fully adaptive time and space discretisation method for phase-field simulation of binary alloy solidification, *Journal of Computational Physics* **225**(2), 1271–1287 (2007).
267. C. Graser and R. Kornhuber, On preconditioned uzawa-type iterations for a saddle point problem with inequality constraints, *Lecture Notes in Computational Science and Engineering* **55**, 91 (2007).
268. J. Rosam, P.K. Jimack and A.M. Mullis, An adaptive, fully implicit multi-grid phase-field model for the quantitative simulation of non-isothermal binary alloy solidification, *Acta Materialia* **56**(17), 4559–4569 (2008).
269. J.R. Green, P.K. Jimack, A.M. Mullis and J. Rosam, Towards a three-dimensional parallel, adaptive, multilevel solver for the solution of non-linear, time-dependent, phase-change problems, *Parallel, Distributed and Grid Computing for Engineering* **21**, 251–274 (2009).
270. A.M. Mullis, J. Rosam and P.K. Jimack, Solute trapping and the effects of anti-trapping currents on phase-field models of coupled thermo-solutal solidification, *Journal of Crystal Growth* **312**(11), 1891–1897 (2010).
271. Z. Guo, J. Mi and P.S. Grant, An implicit parallel multigrid computing scheme to solve coupled thermal-solute phase-field equations for dendrite evolution, *Journal of Computational Physics* **231**(4), 1781–1796 (2012).
272. C. Gräser, R. Kornhuber and U. Sack, Time discretizations of anisotropic Allen-Cahn equations, *IMA Journal of Numerical Analysis* **33**(4), 1226–1244 (2013).
273. C.E. Goodyer, P.K. Jimack, A.M. Mullis, H. Dong and Y. Xie, On the fully implicit solution of a phase-field model for binary alloy solidification in three dimensions, *Advances in Applied Mathematics and Mechanics* **4**(6), 665–684 (2012).
274. L. Vanherpe, F. Wendler, B. Nestler and S. Vandewalle, A multigrid solver for phase field simulation of microstructure evolution, *Mathematics and Computers in Simulation* **80**(7), 1438–1448 (2010), Multiscale modeling of moving interfaces in materials.
275. R. Kornhuber and R. Krause, On multigrid methods for vector-valued Allen-Cahn equations with obstacle potential, *Domain Decomposition Methods in Science and Engineering*, page 307 (2002).
276. G. Amberg, Semisharp phase field method for quantitative phase change simulations, *Physical Review Letters* **91**(26), 265505 (2003).
277. T. Takaki, T. Fukuoka and Y. Tomita, Phase-field simulation during directional solidification of a binary alloy using adaptive finite element method, *Journal of Crystal Growth* **283**(1–2), 263–278 (2005).
278. Y. Li and J. Kim, Phase-field simulations of crystal growth with adaptive mesh refinement, *International Journal of Heat and Mass Transfer* **55**(25–26), 7926–7932 (2012).
279. A. Vondrous, M. Selzer, J. Hötzer and B. Nestler, Parallel Computing for Phase-field Models, *Int. J. High Perform, Comput, Appl.*, **28**(1), 61–72 (February 2014).
280. T. Aoki, S. Ogawa and A. Yamanaka, Multiple-GPU scalability of phase-field simulation for dendritic solidification, *Progress in Nuclear Science and Technology* **2**, 639–642 (2011).
281. R. Altenfeld, M. Apel, Dieteran Mey, B. Böttger, S. Benke and C. Bischof, *OpenMP in the Petascale Era: 7th International Workshop on OpenMP, IWOMP 2011, Chicago, IL, USA, June 13–15, 2011, Proceedings*, chapter Parallelising Computational Microstructure Simulations for Metallic Materials with OpenMP, pages 1–11, Springer Berlin Heidelberg, Berlin, Heidelberg (2011).
282. National Research Council (US), Committee on Integrated Computational Materials Engineering, *Integrated computational materials engineering: a transformational discipline for improved competitiveness and national security*, National Academies Press (2008).

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