Classical density functional theory methods in soft and hard matter

PREFACE

Classical density functional theory methods in soft and hard matter

Guest Editors

Mikko Haataja
Department of Mechanical and Aerospace Engineering, Institute for the Science and Technology of Materials (PRISM) and Program in Applied and Computational Mathematics (PACM), Princeton University, Princeton NJ 08544, USA

László Gránásy
Research Institute for Solid State Physics and Optics, H-1525 Budapest, P.O. Box 49, Hungary and BCAST, Brunel University, Uxbridge, Middlesex, UB8 3PH, UK

Hartmut Löwen
Department of Theoretical Physics, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany

Herein we provide a brief summary of the background, events and results/outcome of the CECAM workshop 'Classical density functional theory methods in soft and hard matter' held in Lausanne between October 21 and October 23 2009, which brought together two largely separately working communities, both of whom employ classical density functional techniques: the soft-matter community and the theoretical materials science community with interests in phase transformations and evolving microstructures in engineering materials. After outlining the motivation for the workshop, we first provide a brief overview of the articles submitted by the invited speakers for this special issue of *Journal of Physics: Condensed Matter*, followed by a collection of outstanding problems identified and discussed during the workshop.

1. Introduction

Classical density functional theory (DFT) is a theoretical framework, which has been extensively employed in the past to study inhomogeneous complex fluids (CF) [1–4] and freezing transitions for simple fluids, amongst other things. Furthermore, classical DFT has been extended to include dynamics of the density field, thereby opening a new avenue to study phase transformation kinetics in colloidal systems via dynamical DFT (DDFT) [5]. While DDFT is highly accurate, the computations are numerically rather demanding, and cannot easily access the mesoscopic temporal and spatial scales where diffusional instabilities lead to complex solidification morphologies. Adaptation of more efficient numerical methods would extend the domain of DDFT towards this regime of particular interest to materials scientists.

In recent years, DFT has re-emerged in the form of the so-called 'phase-field crystal' (PFC) method for solid-state systems [6, 7], and it has been successfully employed to study a broad variety of interesting materials phenomena in both atomic and colloidal systems, including elastic and plastic deformations, grain growth, thin film growth, solid–liquid interface properties, glassy dynamics, nucleation and growth, and diffusive phase transformations at the nano- and mesoscales [8–16]. The appealing feature of DDFT (as applied to solid-state systems) is that it automatically incorporates diffusive dynamics with atomic scale spatial resolution, and it naturally incorporates multiple components, elastic strains, dislocations, free surfaces, and multiple crystalline orientations; all of these features are critical in modeling the behavior of solid-state systems.

Similarities between the problems of interest to the two communities and the complementary nature of the methods they apply suggest that a direct interaction between them should be highly beneficial for both parties. Here we summarize some of the discussions during a three-day CECAM workshop in Lausanne (21–23 October 2009) which was organized in order to bring together researchers from the complex fluids and materials science communities and to foster the exchange of ideas between these two communities. During the course of the

workshop, several open problems relevant to both fields (DFT and PFC) were identified, including developing better microscopically-informed density functionals, incorporating stochastic fluctuations, and accounting for hydrodynamic interactions. The goal of this special issue is to highlight recent progress in DFT and PFC approaches, and discuss key outstanding problems for future work.

The rest of this introductory paper is organized as follows. In section 2, we give a brief overview of the current research topics addressed in this special issue. Then, in section 3, we present a collection of outstanding problems, which have been identified as important for further developments of the two fields and intensely debated at the CECAM workshop. Finally, we close the paper with a few concluding remarks.

2. Research topics addressed in this special issue

This special issue consists of research papers that cover a broad range of interesting subjects, about a half of which are related to the theoretical materials science community and the other half came from the soft-matter community. We begin by discussing papers related to PFC.

Diverse subjects related to the phase-field crystal model include exciting topics such as predicting/controlling the equilibrium phase behavior [19, 18, 17] and kinetics of epitaxial island formation on nano-membranes [20]. Moreover, phase-field crystal modeling has proved to be very successful in simulating homogeneous and heterogeneous crystal nucleation and growth, and several aspects of these phenomena are discussed in this issue [18, 21]. Finally, it is shown how to incorporate additional orientational degrees of freedom within the PFC approach to model liquid crystals [22].

On the DFT side, the other papers in this special issue deal with problems associated with advanced DFT techniques and applications. The existence of a structural instability in sub-critical crystalline fluctuations in a supercooled liquid within a square-gradient theory is discussed in [23]. Fundamental measure theory for hard-body systems is improved by discussing a correction term in detail, as discussed in [24]. A mean-field-like density functional for charges is applied to the effective interaction between charged colloids obtained within a cell model [25]. The remaining articles provide fundamental insight into how to supplement DDFT-type methods with hydrodynamics [26, 27], highlight the role of the projection operator technique in deriving dynamical density functional theories [28], and demonstrate how perturbation methods can be employed to compute the properties of solid–liquid interfaces [29].

This particular collection of papers demonstrates rather convincingly the significant potential that classical density functional techniques possess in modeling complex systems built of either soft or hard matter (or combinations thereof). While the PFC approach offers a simple and appealing means to simulate evolving microstructures in spatially extended system with atomic scale spatial resolution over diffusive time scales, DFT provides both its theoretical underpinning and (hopefully) the means to construct microscopically more quantitative density functionals for use in engineering materials. Outstanding issues within the PFC and DFT approaches, discussed next, will provide further opportunities for interactions between the PFC and DFT communities.

3. Important open issues and exciting avenues for further research

In the following we summarize some of the exciting topics for future research, which were discussed during the CECAM workshop. They concern both fundamental problems and applications, all within the framework of DFT and

PFC. Addressing these issues will provide a framework for future work in these two overlapping fields.

(a) *How to construct a reliable density functional (DF) for soft repulsions?* Most of the recent developments in classical density functional theory were focussed on hard-sphere-like interactions in the framework of fundamental-measure-theory (FMT) [30–33]. While this approach can be extended to additive and nonadditive mixtures [34, 35] and to non-spherical hard objects [36, 37], it is much more difficult to include soft-core interactions, such as inverse-power-law pair-potentials. There have been attempts to include those, mainly using the Ramakrishnan–Yussouff [38] or the weighted-density [39–41] approximation, or other modifications (see e.g., [42, 43]), but the accuracy of these functionals are inferior to that of FMT for hard spheres. Clearly the FMT of Rosenfeld needs an extension for the hard-core Coulomb system. A complementary approach is to start from a density functional for hard orientable objects [36] and to integrate out the orientational degrees of freedom. This would lead to a softened effective repulsion between spherical objects. We mention finally that in the extreme limit of ultrasoft pair potentials, which are penetrable, the mean-field approximation provides a reliable functional [44].

(b) *How to construct a reliable DF beyond perturbation theory?* This is the key to developing accurate, predictive functionals for use in materials science problems. Typically an attractive tail in the interparticle interaction is treated within thermodynamic hard-sphere perturbation theory [45, 46], in most cases at the mean-field level. As this perturbative approach is only justified for weak attraction strengths, there is a great need to go beyond this perturbation theory. A general non-perturbative route, which could be helpful here, is to consider a functional for a mixture and reducing it to an *effective* one-component system. Following this idea, for example effective depletion attractions can be modeled for a one-component system by starting from the binary Asakura–Oosawa functional [34, 35]. This idea still needs to be exploited in a more general sense, i.e. for more general cross-interactions in the mixture. It could also be combined with the idea of using non-spherical hard objects and integrating out the orientational degrees of freedom.

(c) *How to apply the fundamental measure theory to the full phase diagram of lyotropic liquid crystals?* There are already density-functional investigations of liquid–crystal phases of hard spherocylinders [47, 48], but the novel fundamental-measure-theory which was recently proposed for non-spherical objects [36] has never been applied to this problem. In fact, this new functional now needs numerical evaluation for liquid–crystal phases different from isotropic and nematic ones, such as smectic, columnar, plastic crystalline and full orientational ordered crystalline phases [49, 50]. This is mainly a pure numerical resolution problem since the density fields are sharply peaked in the solid phases and need enough grid points, which is at the moment a rather formidable challenge in three spatial dimensions. However, if only orientational degrees of freedoms are considered, the computational effort is greatly reduced; see, e.g., [36, 51, 52].

(d) *The role of fluctuations in DDFT and PFC.* There is a continuing debate about the role of noise in the dynamical density functional theory (see e.g. [53]) and correspondingly also in the phase-field crystal models. Derivations of DDFT from the Smoluchowski level [54] and also within the projection operator technique [5] quite naturally lead to a deterministic equation without any noise. Clearly this is an approximation, which becomes problematic in the

3

vicinity of a critical point or in the case of nucleation problems, where the system has to leave a metastable minimum of the free energy; in the former case, fluctuations are required in order to capture the correct critical behavior (i.e., critical exponents), while in the latter case, fluctuations are needed to establish an escape route of the system from a metastable phase. Other approaches add noise on a more phenomenological level. However, the actual strength of the noise, though fundamentally correlated with the thermal energy, is not known exactly and is treated in most applications as a phenomenological fit parameter; see, e.g., [55, 56]. This problem is a very fundamental one, and, of course, shared by the DDFT and PFC approaches. In more general terms, the addition of noise to the equation of motion in continuum models is not without conceptual difficulties (see [57]), even if noise is properly discretized in the course of the numerical integration. With the noise added, the equilibrium physical properties of the system change. Furthermore, transformation kinetics generally depend on the spatial and temporal steps, and in the limit of infinitely small steps an ultraviolet 'catastrophe' (divergence of the free energy) may occur. Evidently, an 'ultraviolet cut-off', i.e. filtering out the highest frequencies, is required to regularize the unphysical singularity. In the PFC case, a straightforward choice for the cut-off length is the interparticle distance, which is expected to remove the unphysical, small wavelength fluctuations [58, 16, 59, 18]. Perhaps a more elegant way to handle this problem is via renormalizing the model parameters so that with noise one recovers the 'bare' physical properties (see the application of this approach for the Swift–Hohenberg model in [60]). However, further systematic investigations are needed in order to settle this issue.

(e) *The need to clarify the role of the adiabatic approximation.* While DDFT can be derived from more microscopic equations, such as the Smoluchowski equation [54] or the Langevin equations [61] for the individual particles, a major approximation is invoked in the derivation, namely the so-called 'adiabatic approximation'. This approximation assumes that all other observables relax much faster than the one-particle density field [5]. Therefore, the nonequilibrium correlations are replaced by equilibrium ones corresponding to an inhomogeneous reference one-particle density [54]. This enables one to formulate the theory in terms of the time-dependent one-particle density field alone. What is still needed here is a more general theory which provides the next-leading order beyond the adiabatic approximation. This improved theory would not only provide more fundamental insight into the DDFT itself; it would also pave the way to many applications where the simple DDFT fails.

(f) *How to apply and exploit DDFT for active matter?* The collective behavior of self-propelled particles with internal driving motors is a topic of active research [62, 63]. Given that the particle dynamics can be described in terms of driven Brownian motion, a dynamical density functional theory can be derived in a straightforward manner. In a first application, DDFT was employed to describe aggregation phenomena near system boundaries for driven rod-like colloidal particles [64]. The potential of DDFT for 'active' particles should be exploited more in the future, as it provides a microscopic approach to investigate nonequilibrium effects, such as swarming and jamming.

(g) *How to construct a PFC model for inhomogeneous liquid crystals?* The traditional PFC model [6, 7] describes a two-dimensional one-component solid phase by a single inhomogeneous sinusoidal density field. The PFC approach has been generalized to mixtures by including more than a single

density field [11] and to anisotropic particles with a fixed orientation [65]. However, it has never been applied to liquid crystals which are made by particles with intrinsic *orientational degrees of freedom.* Based on discussion during the CECAM workshop, a link towards the PFC model has been elaborated and the corresponding PFC model for liquid crystals was derived, see article [22] in this special issue. The extended PFC model contains both the translational density and the local orientational degree of ordering as well as a local director field. The model exhibits stable isotropic, nematic, smectic A, columnar, plastic crystalline and orientationally ordered crystalline phases and bears therefore much richer phases than the original PFC. A large-scale numerical exploration of this PFC model still needs to be performed. The derivation exploits the connection between DDFT and PFC, which was highlighted in [66] for spherical particles, and is based on recent generalizations of DDFT to rod-like Brownian particles [67, 64].

(h) *How to incorporate hydrodynamic interactions between particles in dense driven systems of colloids?* In dense colloidal dispersions, hydrodynamic interactions between the particles play a major role in their collective behavior. While these interactions affect neither structural correlations nor the equilibrium phase behavior, they have a profound effect on the dynamics both in equilibrium and non-equilibrium [68]. Recently, DDFT was extended to include hydrodynamic interactions on the pairwise level of the mobility tensors [69]. This kind of DDFT needs more applications as well as a fundamental development towards higher-order mobility tensors beyond the pairwise level or to a description, which includes lubrication forces between colloidal particles at small interparticle separations.

(i) *How to systematically construct effective, low-frequency representations from DFT/DDFT?* Given an accurate and predictive density functional, which incorporates interaction potentials between the constituent species in a multi-component system, building an effective description would be highly desirable as it would provide an alternative to purely atomistic approaches (e.g., molecular dynamics simulations) and enable the simulation of quantitative, microscopically-informed, continuum systems across diffusive time scales. The first challenge, of course, is the development of such functionals, as already discussed in item (b) above. Once this challenge has been overcome, the next step would be to project out the dynamics of the relevant degrees of freedom from the full DDFT description. Physically, one would expect that the shape of a single peak in the density would relax much faster than, say, the distance between peak centers. Therefore, it should be possible to 'slave' the high-frequency modes associated with the peak shapes to the more slowly evolving modes with low spatial frequencies.

(j) *How to build numerically efficient, quantitative PFC models for a broad spectrum of metallic materials?* Viewed as an extension of the traditional phase-field method (see, e.g., [70–74] for comprehensive reviews), PFC incorporates microscopic physics (crystal symmetry, grain orientation, topological defects) in a phenomenological manner. A practical issue in numerically integrating the dynamic PFC equation is that the grid spacing is constrained to be a fraction of the lattice spacing (typically $\Delta x \sim a/8$), making large-scale simulations challenging in three spatial dimensions. It is thus highly desirable to develop a methodology that would allow one to tune important materials parameters such as crystal symmetry, lattice spacing, elastic constants, surface energies and stresses, dislocation core energy, and dislocation mobility, without sacrificing numerical efficiency. The issue of constructing PFC free energies, which give rise to a given crystal symmetry,

has been addressed very recently; see, e.g., [17–19]. Going beyond the question of crystal symmetry, an appealing possibility is to further develop the so-called amplitude equation approach [75–77], in which the density field is essentially expressed in terms of slowly-varying envelope functions (i.e., amplitudes), modulated by the fundamental spatial periodicity of the density. In fact, it has been demonstrated recently that such an approach provides a truly multi-scale approach to studying phase transformations in solid–liquid systems [78]. The goal is to construct amplitude equations, which accurately incorporate, e.g., surface tension anisotropies for simulations of solid–solid, solid–liquid, and solid–vapor systems. Alternatively, one can work directly with the PFC density field and introduce additional model parameters which can be fitted so that a required set of physical properties is recovered, such as the properties of the solid–liquid interface in pure iron [79].

(k) *How to simulate electronic materials with PFC?* Ferroelectrics comprise an interesting class of materials, which undergo a structural phase transformation (typically cubic-to-tetragonal) below a Curie temperature and acquire a non-zero electric polarization. It has been suggested that the manipulation of these polarization domains by means of an external field can be exploited in novel non-volatile memory devices [80, 81]. The PFC approach would present an appealing means to study ferroelectrics exhibiting one or more (ferroic) order parameters, provided that the crystal lattice can be coupled to the local order parameter(s) in a physically-based manner.

4. Concluding remarks

The workshop 'Classical density functional theory methods in soft and hard matter' has established the first contact between the soft-matter community working with advanced classical density functional techniques and a theoretical materials science community working with engineering materials and armed with a simple but numerically very efficient dynamical density functional technique, the phase-field crystal method. A large number of common problems have been identified, which represent challenges for both communities during the coming years. This has been borne out by the lively discussions and some of the provocative talks. The organizers think that the workshop proved to be a truly successful event, matching to the high standards of the CECAM workshops, and hope that the workshop will indeed catalyze a long-term interaction between the two communities.

As a final note, we would like to emphasize that progress in the areas highlighted in this special issue will positively impact both fields, and we expect that these issues will provide the natural link for collaborations and intellectual exchanges between these traditionally separate-yet-allied fields. In particular, such activities would lead to significant improvements in the applicability and versatility of classical DFT methods in both soft and hard matter systems, for the common benefit of physicists, chemists, and materials scientists.

References

[1] Evans R 1979 *Adv. Phys.* **28** 143
[2] Oxtoby D W 1991 *Liquids, Freezing and the Glass Transition (Session LI (1989) of Les Houches Summer Schools of Theoretical Physics)* (Amsterdam: North Holland) p 147
[3] Singh Y 1991 *Phys. Rep.* **207** 351
[4] Löwen H 1994 *Phys. Rep.* **237** 249
[5] Español P and Löwen H 2009 *J. Chem. Phys.* **131** 244101
[6] Elder K R, Katakowski M, Haataja M and Grant M 2002 *Phys. Rev. Lett.* **88** 245701
[7] Elder K R and Grant M 2004 *Phys. Rev.* E **70** 051605

[8] Berry J, Grant M and Elder K R 2006 *Phys. Rev.* E **73** 031609
 [9] Stefanovic P, Haataja M and Provatas N 2006 *Phys. Rev. Lett.* **96** 225504
[10] Wu K-A and Karma A 2007 *Phys. Rev.* B **76** 184107
[11] Elder K R, Provatas N, Berry J, Stefanovic P and Grant M 2007 *Phys. Rev. B* **75** 064107
[12] Berry J, Elder K R and Grant M 2008 *Phys. Rev.* E **77** 061506
[13] Huang Z-F and Elder K R 2008 *Phys. Rev. Lett.* **101** 158701
[14] Wu K-A and Voorhees P W 2009 *Phys. Rev.* B **80** 125408
[15] Stefanovic P, Haataja M and Provatas N 2009 *Phys. Rev.* E **80** 046107
[16] Tegze G, Gránásy L, Tóth G I, Podmaniczky F, Jaatinen A, Ala-Nissial T and Pusztai T 2009
      Phys. Rev. Lett. **103** 035702
[17] Jaatinen A and Ala-Nissila T 2010 Extended phase diagram of the three-dimensional phase
      field crystal model *J. Phys.: Condens. Matter* **22** 205402
[18] Tóth G I, Tegze G, Pusztai T, Tóth G and Gránásy L 2010 Polymorphism, crystal nucleation
      and growth in the phase-field crystal model in 2D and 3D *J. Phys.: Condens. Matter* **22**
      364101
[19] Wu K-A, Plapp M and Voorhees P 2010 Controlling crystal symmetries in phase-field crystal
      models *J. Phys.: Condens. Matter* **22** 364102
[20] Elder K R and Huang Z-F 2010 A phase field crystal study of epitaxial island formation on
      nanomembranes *J. Phys.: Condens. Matter* **22** 364103
[21] Backofen R and Voigt A 2010 A phase-field-crystal approach to critical nuclei *J. Phys.:
      Condens. Matter* **22** 364104
[22] Löwen H 2010 A phase-field-crystal model for liquid crystals *J. Phys.: Condens. Matter* **22**
      364105
[23] Harrowell P 2010 On the existence of a structural instability in sub-critical crystalline
      fluctuations in a supercooled liquid *J. Phys.: Condens. Matter* **22** 364106
[24] Hansen-Goos H and Mecke K 2010 Tensorial density functional theory for non-spherical
      hard-body fluids *J. Phys.: Condens. Matter* **22** 364107
[25] Denton A R 2010 Poisson–Boltzmann theory of charged colloids: limits of the cell model for
      salty suspensions *J. Phys.: Condens. Matter* **22** 364108
[26] Rauscher M 2010 DDFT for Brownian particles and hydrodynamics *J. Phys.: Condens.
      Matter* **22** 364109
[27] Marini Bettolo Marconi U and Melchionna S 2010 Dynamic density functional theory versus
      kinetic theory of simple fluids *J. Phys.: Condens. Matter* **22** 364110
[28] Majaniemi S, Provatas N and Nonomura M 2010 Effective model hierarchies for dynamic and
      static classical density functional theories *J. Phys.: Condens. Matter* **22** 364111
[29] Warshavsky V B and Song X 2010 Perturbation theory for solid–liquid interfacial free
      energies *J. Phys.: Condens. Matter* **22** 364112
[30] Rosenfeld Y, Schmidt M, Löwen H and Tarazona P 1997 *Phys. Rev.* E **55** 4245
[31] Roth R, Evans R, Lang A and Kahl G 2002 *J. Phys: Condens. Matter* **14** 12063
[32] Tarazona P, Cuesta J A and Martinez-Raton Y 2008 *Density Functional Theories of Hard
      Particle Systems* (*Springer Lecture Notes in Physics* vol 753) (Berlin: Springer) p 247
[33] Roth R 2010 *J. Phys.: Condens. Matter* **22** 063102
[34] Schmidt M, Löwen H, Brader J M and Evans R 2000 *Phys. Rev. Lett.* **85** 1934
[35] Schmidt M, Löwen H, Brader J M and Evans R 2002 *J. Phys.: Condens. Matter* **14** 9353
[36] Hansen-Goos H and Mecke K 2009 *Phys. Rev. Lett.* **102** 018302
[37] Esztermann A, Reich H and Schmidt M 2006 *Phys. Rev.* E **73** 011409
[38] Ramakrishnan T V and Yussouff M 1979 *Phys. Rev.* B **19** 2775
[39] Denton A R and Ashcroft N W 1989 *Phys. Rev.* A **39** 4701
[40] Hasegawa M 1994 *J. Phys. Soc. Japan* **63** 2215
[41] Kol A and Laird B B 1997 *Mol. Phys.* **90** 951
[42] van Teeffelen S, Löwen H and Likos C N 2008 *J. Phys.: Condens. Matter* **20** 404217
[43] van Teeffelen S, Hoffmann N, Likos C N and Löwen H 2006 *Europhys. Lett.* **75** 583
[44] Likos C N, Hoffmann N, Löwen H and Louis A A 2002 *J. Phys.: Condens. Matter* **14** 7681
[45] Curtin W A and Ashcroft N W 1986 *Phys. Rev. Lett.* **56** 2775
[46] Likos C N, Németh Z T and Löwen H 1994 *J. Phys.: Condens. Matter* **6** 10965
[47] Poniewierski A and Holyst R 1988 *Phys. Rev. Lett.* **61** 2461
[48] Graf H and Löwen H 1999 *J. Phys.: Condens. Matter* **11** 1435
[49] Bolhuis P and Frenkel D 1997 *J. Chem. Phys.* **106** 666
[50] Frenkel D, Mulder B M and McTague J P 1984 *Phys. Rev. Lett.* **52** 287
[51] Härtel A and Löwen H 2010 *J. Phys.: Condens. Matter* **22** 104112
[52] Härtel A, Blaak R and Löwen H 2010 Towing, breathing, splitting, and overtaking in driven
      colloidal liquid crystals *Phys. Rev.* E **81** 051703
[53] Archer A J and Rauscher M 2004 *J. Phys. A: Math. Gen.* **37** 9325
[54] Archer A J and Evans R 2004 *J. Chem. Phys.* **121** 4246
[55] Ramos J A P, Granato E, Achim C V, Ying S C, Elder K R and Ala-Nissila T 2008 *Phys. Rev.*
      E **78** 031109

7

[56] Hubert J, Cheng M and Emmerich H 2009 *J. Phys.: Condens. Matter* **21** 464108
[57] Plapp M 2010 *Philos. Mag.* submitted
[58] Pusztai T, Tegze G, Tóth G I, Környei L, Bansel G, Fan Z and Gránásy L 2008 *J. Phys.: Condens. Matter* **20** 404205
[59] Tegze G, Bansel G, Tóth G I, Pusztai T, Fan Z and Gránásy L 2009 *J. Comput. Phys.* **228** 1612
[60] Gross N A, Ignatiev M and Chakraborty B 2000 *Phys. Rev. E* **62** 6116
[61] Marconi V M B and Tarazona P 2000 *J. Phys.: Condens. Matter* **12** A413
[62] Toner J, Tu Y and Ramaswamy S 2005 *Ann. Phys.* **318** 170
[63] Lauga E and Powers T R 2009 *Rep. Prog. Phys.* **72** 096601
[64] Wensink H H and Löwen H 2008 *Phys. Rev. E* **78** 031409
[65] Prieler R, Hubert J, Li D, Verleye B, Haberkern R and Emmerich H 2009 *J. Phys.: Condens. Matter* **21** 464110
[66] van Teeffelen S, Backofen R, Voigt A and Löwen H 2009 *Phys. Rev. E* **79** 051404
[67] Rex M, Wensink H H and Löwen H 2007 *Phys. Rev. E* **76** 021403
[68] Dhont J K G 1996 *An Introduction to Dynamics of Colloids* (Amsterdam: Elsevier)
[69] Rex M and Löwen H 2008 *Phys. Rev. Lett.* **101** 148302
[70] Elder K R, Grant M, Provatas N and Kosterlitz J M 2001 *Phys. Rev. E* **64** 021604
[71] Chen L Q 2002 *Annu. Rev. Mat. Res.* **32** 113
[72] Boettinger W J, Warren J A, Beckermann C and Karma A 2002 *Annu. Rev. Mat. Res.* **32** 163
[73] Gránásy L, Pusztai T and Warren J A 2004 *J. Phys.: Condens. Matter* **16** R1205
[74] Singer-Loginova I and Singer H M 2008 *Rep. Prog. Phys.* **71** 106501
[75] Goldenfeld N, Athreya B P and Dantzig J A 2005 *Phys. Rev. E* **72** 020601
[76] Yeon D-H, Huang Z-F, Elder K R and Thornton K 2010 *Phil. Mag.* **90** 237
[77] Elder K R, Huang Z-F and Provatas N 2010 *Phys. Rev. E* **81** 011602
[78] Athreya B P, Goldenfeld N, Dantzig J A, Greenwood M and Provatas N 2007 *Phys. Rev. E* **76** 056706
[79] Jaatinen A, Achim C V, Elder K R and Ala-Nissila T 2009 *Phys. Rev. E* **80** 031602
[80] Chu M-W *et al* 2004 *Nat. Mater.* **3** 87
[81] Rudiger A and Waser J 2008 *J. Alloy Compounds* **449** 2