

promoting access to White Rose research papers



Universities of Leeds, Sheffield and York
<http://eprints.whiterose.ac.uk/>

This is an author produced version of an article published in **arXiv**.

White Rose Research Online URL for this paper:

<http://eprints.whiterose.ac.uk/76214/>

Published article:

Archer, AJ, Rucklidge, AM and Knobloch, E (2013) *Formation of quasicrystals and other exotic crystal structures in a soft-core fluid: a tale of two length scales.*

Formation of quasicrystals and other exotic crystal structures in a soft-core fluid: a tale of two length scales

A.J. Archer*, A.M. Rucklidge† and E. Knobloch#

**Department of Mathematical Sciences, Loughborough University, Loughborough LE11 3TU, UK*

†*Department of Applied Mathematics, University of Leeds, Leeds LS2 9JT, UK*

#*Department of Physics, University of California at Berkeley, Berkeley, CA 94720, USA*

A two-dimensional system of soft particles interacting via a two-length-scale potential is studied. Density functional theory reveals the existence of a fluid phase and two crystalline phases with different lattice spacing. Of these the larger lattice spacing crystalline phase takes the form of a ‘crystal liquid’ state with a small fraction of mobile particles. Near the transition between this phase and the smaller lattice spacing crystalline phase, quasicrystalline structures may be created by a competition between linear instability at one scale and nonlinear selection of the other. The results are confirmed using Brownian dynamics simulations.

PACS numbers: 61.50.Ah, 61.44.Br, 05.20.-y, 64.70.D-

Regular crystals are ordered arrangements of atoms or molecules with rotation and translation symmetries. Quasicrystals, discovered in 1984 [1], lack the lattice symmetries of crystals and yet have discrete Fourier spectra. Quasicrystals have been found not only in metals but also in systems of nanoparticles [2], mesoporous silica [3], and soft-matter systems [4]. The latter include micellar melts [5, 6] formed, e.g., from linear, dendrimer or star block copolymers, comprising a stiff hydrophobic polymer core surrounded by a corona of flexible hydrophilic polymer chains. The main theoretical approach to investigating the stability of metallic or micellar quasicrystals involves minimising an appropriate energy, but the principle underlying their stability is not known [7, 8].

Patterns with quasicrystalline structure, or quasipatterns, were discovered in Faraday wave experiments in the 1990s [9, 10]. Many quasipatterns are characterised by two length scales [11, 12], and recent work suggests that nonlinear interaction between these scales can stabilise such patterns [13, 14] and this is so for soft-matter quasicrystals as well [15–17]. In this Letter, we explore this mechanism in detail, by considering a system of particles interacting via a simple potential with two length scales. We compute the phase diagram, finding crystals of each length scale and a region of metastable quasicrystals with both length scales prominent in their Fourier spectra. These form in a region where the linear growth of density fluctuations in a quenched uniform fluid favors one length scale but nonlinear stability favors the other.

The effective ‘coarse-grained’ interaction potentials between the centres of mass of polymers, dendrimers or other such macromolecules, are soft. By this we mean that they are finite for all separation distances r , because the centre of mass of such soft objects does not necessarily coincide with any individual monomer. The soft effective pair potential between such particles can be approximated as $V(r) = \epsilon e^{-(r/R)^n}$. Simple linear polymers correspond to the case $n = 2$ with the length R of order the radius of gyration and the energy ϵ for such a

pair of polymers to fully overlap of order $2k_B T$, where k_B is Boltzmann’s constant and T is the temperature [18–28]. Dendrimers, due to the nature of their chemical architecture, can have an effective interaction with a higher value of n ; such systems form so-called ‘cluster crystals’ [28] and there has been a great deal of interest in soft potential models for these systems [29–43].

Here we consider a model two-dimensional system of soft ‘particles’ that interact via the potential

$$V(r) = \epsilon e^{-(r/R)^8} + \epsilon A e^{-(r/R_s)^8}. \quad (1)$$

The energy for complete overlap is $(1 + A)\epsilon$. The most important feature of this potential is that it has a ‘shoulder’ when the dimensionless parameter $A \neq 0$, with two length scales. The radius of the core is R and the radius of the shoulder is $R_s > R$. Such a potential is a simple coarse-grained model for the effective interaction between polymers or micelles formed, e.g., from linear, dendrimer or star block copolymers, which have a stiff hydrophobic core surrounded by a corona of flexible hydrophilic chains. A related, piecewise constant potential is used in Ref. [17]. In the following we set the dimensionless interaction energy parameter $\beta\epsilon = 1$, where $\beta = (k_B T)^{-1}$, and fix the ratio of the two length scales to be $R_s/R = 1.855$.

We use density functional theory (DFT) [44–46] to study this system. The grand free energy is

$$\begin{aligned} \Omega[\rho(\mathbf{r})] = & k_B T \int d\mathbf{r} \rho(\mathbf{r}) [\ln \Lambda^2 \rho(\mathbf{r}) - 1] \\ & + \mathcal{F}_{ex}[\rho(\mathbf{r})] + \int d\mathbf{r} (\Phi(\mathbf{r}) - \mu) \rho(\mathbf{r}), \quad (2) \end{aligned}$$

which is a functional of the one-body (number) density of the particles, $\rho(\mathbf{r})$, where $\mathbf{r} = (x, y)$. The first term is the ideal-gas contribution to the free energy, Λ is the (irrelevant) thermal de Broglie wavelength, μ is the chemical potential, $\Phi(\mathbf{r})$ is any external potential that may be confining the system and $\mathcal{F}_{ex}[\rho(\mathbf{r})]$ is the excess Helmholtz

free energy from the interactions between the particles. The equilibrium density distribution is that which minimises $\Omega[\rho(\mathbf{r})]$; the corresponding minimum is the thermodynamic grand potential of the system. For a system in the bulk fluid state (i.e., where $\Phi(\mathbf{r}) \equiv 0$), the minimising density is uniform, $\rho = \rho_0$. However, for other state points, when the system freezes to form a solid, Ω is minimised by nonuniform density distributions, exhibiting sharp peaks. For the systems of soft-core particles considered here, one may approximate \mathcal{F}_{ex} as [18]:

$$\mathcal{F}_{ex}[\rho(\mathbf{r})] = \frac{1}{2} \int d\mathbf{r} \int d\mathbf{r}' \rho(\mathbf{r}) V(|\mathbf{r} - \mathbf{r}'|) \rho(\mathbf{r}'). \quad (3)$$

This functional generates the random phase approximation (RPA) for the pair direct correlation function $c^{(2)}(\mathbf{r}, \mathbf{r}') \equiv -\beta \frac{\delta^2 \mathcal{F}_{ex}}{\delta \rho(\mathbf{r}) \delta \rho(\mathbf{r}')} = -\beta V(|\mathbf{r} - \mathbf{r}'|)$ [44–46]. If we assume that these are Brownian particles with dynamics governed by

$$\dot{\mathbf{r}}_i = -\Gamma \nabla_i U(\{\mathbf{r}_i\}, t) + \Gamma \mathbf{X}_i(t), \quad (4)$$

where the index $i = 1, \dots, N$ labels the particles, $U(\{\mathbf{r}_i\}, t) = \sum_{i=1}^N \Phi(\mathbf{r}_i) + \sum_{i \neq j} V(\mathbf{r}_i - \mathbf{r}_j)$ is the potential energy of the system and $\mathbf{X}_i(t)$ is a white noise term, we can investigate the dynamics of the system using Dynamic Density Functional Theory (DDFT) [47–50] in the form

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} = \Gamma \nabla \cdot \left[\rho(\mathbf{r}, t) \nabla \frac{\delta \Omega[\rho(\mathbf{r}, t)]}{\delta \rho(\mathbf{r}, t)} \right], \quad (5)$$

where $\rho(\mathbf{r}, t)$ is now the time-dependent nonequilibrium one-body density profile and $\Gamma \equiv \beta D$ is the mobility. Here D is the diffusion coefficient. In deriving this theory we have used the equilibrium Helmholtz free energy functional \mathcal{F} to approximate the unknown nonequilibrium free energy.

Fig. 1(a) shows the equilibrium phase diagram calculated using Picard iteration of the DFT Euler–Lagrange equation, starting either from the profile for a nearby state point or a uniform density profile with a small random value added to each point. As the fluid density is increased, the system freezes to form one of two distinct solid phases (Fig. 2): for larger values of A the system forms a hexagonal crystal with a large lattice spacing (referred to as ‘crystal A’), but for smaller values of A it forms a hexagonal crystal with a much smaller lattice spacing (‘crystal B’). The red regions in the diagram denote thermodynamic coexistence between two different phases at the same temperature, pressure and chemical potential.

To understand the phase diagram we study the structure and stability of a uniform liquid with density ρ_0 and $\Phi(\mathbf{r}) \equiv 0$. We follow [44, 49, 51, 52] and expand Eq. (5) in powers of $\tilde{\rho}(\mathbf{r}, t) \equiv \rho(\mathbf{r}, t) - \rho_0$. Retaining only linear terms, we find that the growth/decay of different Fourier modes of wave number k follows

$\hat{\rho}(k, t) = \hat{\rho}(k, 0) \exp[\omega(k)t]$, where $\omega(k)$ satisfies the dispersion relation [49, 52]

$$\omega(k) = -\Gamma k_B T k^2 (1 - \rho_0 \hat{c}(k)). \quad (6)$$

Here $\hat{c}(k)$ is the Fourier transform of the pair direct correlation function; within RPA $\hat{c}(k) = -\beta \hat{V}(k)$, where $\hat{V}(k)$ is the Fourier transform of the pair potential in Eq. (1). In an equilibrium fluid the static structure factor $S(k) \equiv (1 - \rho_0 \hat{c}(k))^{-1} > 0$ for all values of k ; such a fluid is therefore stable [53]. Within RPA the two length scales in the pair potential lead, for certain ranges of parameter values, to a static structure factor $S(k)$ with two peaks. Fig. 1(b)–(d) shows that as A increases the smaller wave number peak in $S(k)$ grows and comes to dominate the larger wave number peak. Fig. 1(e)–(g) shows analogous behavior of the dispersion relation at several points in or on the boundary of the linearly unstable region $\omega(k_{max}) = 0$ where k_{max} is the wave number of the *higher* peak (blue dashed line in Fig. 1(a)): as A increases the instability shifts from large wave number (Fig. 1(g)) to small wave number (Fig. 1(e)). The short and long length scales are simultaneously marginally stable at $A = 1.067$ and $\rho_0 R^2 = 2.95$ (Fig. 1(f)); this point lies on the pink dotted line in Fig. 1(a) corresponding to a pair of equal height peaks in the dispersion relation. Above (below) this line, the peak at smaller (larger) wave number k is higher, indicating that the longer (shorter) length scale density fluctuations grow the fastest. The black double dotted lines indicate the location of $\omega(k_{max}) = 0$ for the *lower* peak in the dispersion relation. When the system is quenched from a stable liquid state to a state point with density ρ_0 above the blue dashed line, certain wave numbers will grow as described by the dispersion relation (6).

Fig. 2, third panel in the top row, shows the density profile of the larger lattice spacing crystal A phase for a state point not far from the transition to the smaller lattice spacing crystal B phase. However, the panel below displaying $\log[R^2 \rho(\mathbf{r})]$ reveals an interconnected network of ‘channels’ between the density peaks. The particles contributing to this part of the density profile are fluid in the sense that they can move freely throughout the whole system, unlike the majority of the particles that are located in the density peaks fixed on the lattice sites. We refer to this structure as the ‘crystal liquid’ state. This state minimises the free energy for $A > A_{co}$, where A_{co} is the value at coexistence, and is reminiscent of the way electrons arrange themselves in a metal, where the vast majority are bound on the lattice sites to the metal atomic nuclei. However, there are some, the conduction band electrons, that are free to move throughout the crystal. The crucial differences between the present system and a metal are that (i) the present particles are classical, and (ii) they *self-organise* to form this remarkable structure – there is no equivalent of the atomic nuclei to provide a periodic array of potential wells into which the majority of the particles condense.

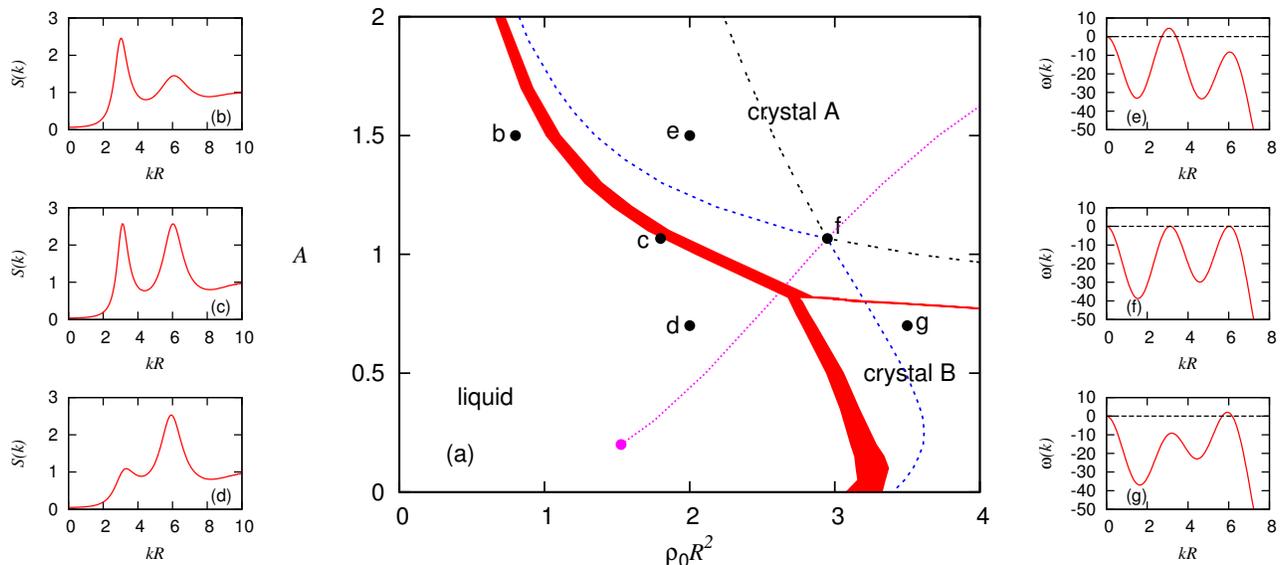


FIG. 1: (color online) Phase diagram, static structure factor $S(k)$ and dispersion relation $\omega(k)$ for $\beta\epsilon = 1$ and $R_s/R = 1.855$. (a) The bulk system phase diagram in the $(\rho_0 R^2, A)$ plane. The system exhibits a uniform fluid phase and two crystal phases: the larger lattice spacing ‘crystal A’ phase and the smaller lattice spacing ‘crystal B’ phase. The regions filled in red denote areas where there is two-phase coexistence between the different phases. The blue dashed line denotes the linear instability threshold for the liquid phase while the pink dotted line terminating in a circle is the locus where the two peaks in the dispersion relation (6) have the same height. The circle denotes the point where the smaller- k peak disappears. (b)–(d) $S(k)$ for (b) $(\rho_0 R^2, A) = (0.8, 1.5)$, (c) $(1.8, 1.067)$, (d) $(2, 0.7)$. (e)–(g) $\omega(k)$ for (e) $(\rho_0 R^2, A) = (2, 1.5)$, (f) $(2.95, 1.067)$, (g) $(3.5, 0.7)$.

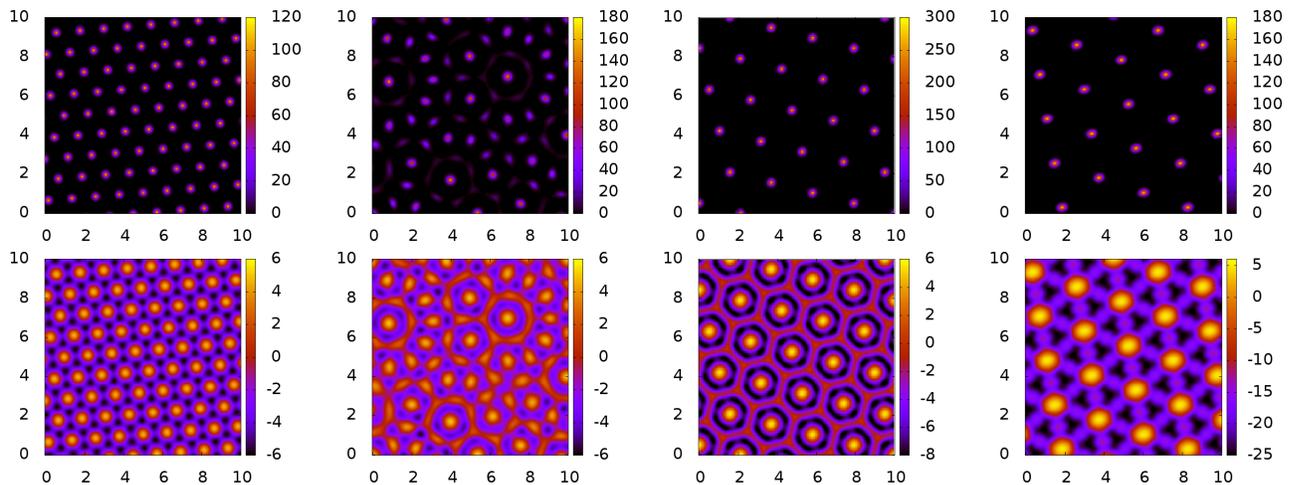


FIG. 2: (color online) Density profiles from DFT showing $R^2\rho(\mathbf{r})$ (upper panels) and $\log[R^2\rho(\mathbf{r})]$ (lower panels). From left to right: $(\rho_0 R^2, A) = (4.4, 0.7)$ (typical of the small length scale crystal ‘crystal B’), $(3.5, 0.76)$, $(4.0, 0.8)$ (both near the transition from ‘crystal A’ to ‘crystal B’) and $(2.7, 2)$ (typical of the large length scale crystal ‘crystal A’). The second set of panels shows quasicrystalline ordering with numerous defects, while the third set reveals a network of connected density, indicating that the particles in this part of the crystal are fluid, and able to move throughout the system. There are also similar connected fragments in the disordered $(3.5, 0.76)$ profile, but because of the disorder, these do not percolate the system.

As A decreases towards A_{co} , the fraction of particles in this ‘conduction band’ increases and in fact it is this growing number that triggers the formation of the smaller length scale crystal: these mobile particles freeze to form the extra peaks of crystal B.

To confirm the existence of the ‘crystal liquid’ state we calculated the density profile for a system confined within a square confining potential Φ of size $L \times L$ with hard walls, and compared the results with Brownian dynamics (BD) simulations, i.e., simulations of N particles

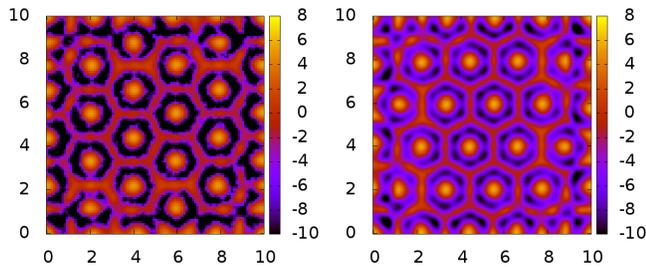


FIG. 3: $\log[R^2\rho(\mathbf{r})]$ for a system of $N = 600$ particles with $(\rho_0 R^2, A) = (4.0, 0.8)$ confined in a square region of side $L = 10R$. The left panel displays the BD simulation result, while the right panel is from DFT. The system forms crystal A with a density profile consisting of an array of peaks surrounded by a connected network within which the particles are free to move – the ‘crystal liquid’ phase.

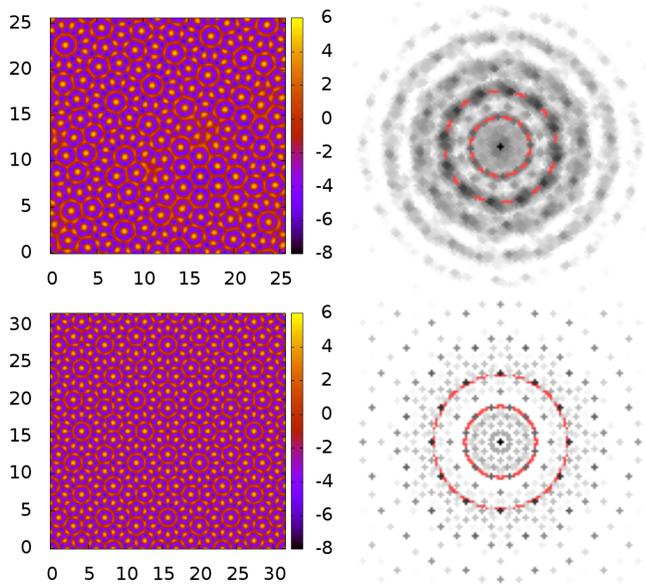


FIG. 4: Left: $\log[R^2\rho(\mathbf{r})]$ from DFT, for $(\rho_0 R^2, A) = (3.5, 0.8)$. Right: the corresponding Fourier transforms. The 12-fold symmetry is indicative of quasicrystalline ordering. The upper density profile was obtained from random initial conditions, while the lower one was started from initial conditions with quasicrystalline symmetry.

evolving according to Eq. (4). Averaging over the positions of the particles to calculate the density profile, we find remarkably good agreement between the DFT and the BD results (Fig. 3).

Formation of a ‘quasicrystal’: A striking aspect of the phase diagram in Fig. 1(a) is that the phase transition between the two different crystal phases (thin red region) is well away from where the two peaks in the dispersion relation have the same height (pink dotted line). A uniform system quenched to the region above the coexistence of the two crystal phases but below this line will initially generate small length scale density fluctuations and the

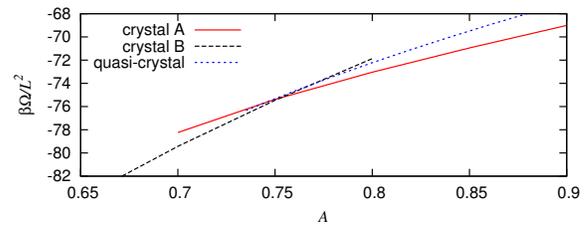


FIG. 5: Grand potential Ω per unit area for $\beta\mu = 39$ as a function of A for the two different crystal structures together with the free energy for the quasicrystalline solution displayed in Fig. 4. There is a point where all three have almost the same free energy, but the quasicrystal solution never corresponds to the global minimum of the free energy. The crystal A phase is of ‘crystal liquid’ type throughout the range of A shown.

system behaves as if it were going to form crystal B. However, the true minimum of the free energy is the larger length scale crystal. Thus, as growing density fluctuations reach the nonlinear regime, the system seeks to go to the longer length scale structure but the smaller length scale imprinted from the linear growth regime leads to frustration. Sometimes the system is able to evolve to the larger length scale crystal; at other times it stays stuck in the metastable small length scale crystal B structure. However, often the system forms a state with density peaks on both length scales, but no long range order. In Fig. 4 we display two rather striking density profiles calculated at a state point in this region of the phase diagram. The upper density profile was calculated using Picard iteration starting from random initial conditions. The density profile has many defects, but it has definite quasicrystalline ordering, as can be seen from the corresponding Fourier transform. The lower panels in Fig. 4 show a defect-free quasicrystal approximant, started from carefully chosen initial conditions. The two wavenumbers $k_1 R = 3.2$ and $k_2 R = 6.0$ corresponding to the maxima in $\omega(k)$ are indicated in the Fourier transforms.

The Picard iteration of the Euler–Lagrange equation corresponds to fictitious dynamics since it does not conserve the total number of particles in the system, $N \equiv \int d\mathbf{r}\rho(\mathbf{r})$. The true dynamics is governed by the DDFT equation (5). Evolving this equation can be rather slow but in most cases the same qualitative behavior is observed, although the system sometimes gets stuck in the smaller length scale crystal B that initially forms in the linear growth regime. This is a consequence of the conserved dynamics. For $\beta\epsilon = 1$, $R_s/R = 1.855$ the quasicrystals we find are never the minimum free energy state (Fig. 5). The quasicrystalline state in Fig. 4 remains stable for $1.77 < R_s/R < 2.18$, but we have not calculated the full phase diagram for $R_s \neq 1.855$. We believe it may be possible to use nonlinear dynamics techniques [14] to compute the stability properties of these states by reducing the DDFT description in Eq. (5) to a phase field

crystal model, cf. [52, 54–57]. We expect that the observed quasicrystal formation mechanism (linear growth of one length scale, but nonlinear selection favoring another) may well apply more generally.

Acknowledgement: This work was supported in part by the National Science Foundation under grant DMS-1211953 (EK). We are grateful for discussions with R. Lifshitz and P. Olmsted.

-
- [1] D. Shechtman, I. Blech, D. Gratias, and J. W. Cahn, *Phys. Rev. Lett.* **53**, 1951 (1984).
- [2] D. V. Talapin, E. V. Shevchenko, M. I. Bodnarchuk, X. C. Ye, J. Chen, and C. B. Murray, *Nature* **461**, 964 (2009).
- [3] C. H. Xiao, N. Fujita, K. Miyasaka, Y. Sakamoto, and O. Terasaki, *Nature* **487**, 349 (2012).
- [4] T. Dotera, *Isr. J. Chem.* **51**, 1197 (2011).
- [5] X. B. Zeng, G. Ungar, Y. S. Liu, V. Percec, S. E. Dulcey, and J. K. Hobbs, *Nature* **428**, 157 (2004).
- [6] S. Fischer, A. Exner, K. Zielske, J. Perlich, S. Deloudi, W. Steurer, P. Lindner, and S. Forster, *Proc. Nat. Acad. Sci. USA* **108**, 1810 (2011).
- [7] A. S. Keys and S. C. Glotzer, *Phys. Rev. Lett.* **99**, 235503 (2007).
- [8] C. R. Iacovella, A. S. Keys, and S. C. Glotzer, *Proc. Nat. Acad. Sci. USA* **108**, 20935 (2011).
- [9] B. Christiansen, P. Alstrom, and M. T. Levinsen, *Phys. Rev. Lett.* **68**, 2157 (1992).
- [10] W. S. Edwards and S. Fauve, *Phys. Rev. E* **47**, R788 (1993).
- [11] W. B. Zhang and J. Viñals, *J. Fluid Mech.* **336**, 301 (1997).
- [12] R. Lifshitz and D. M. Petrich, *Phys. Rev. Lett.* **79**, 1261 (1997).
- [13] A. M. Rucklidge and M. Silber, *SIAM J. Appl. Dynam. Syst.* **8**, 298 (2009).
- [14] A. M. Rucklidge, M. Silber, and A. C. Skeldon, *Phys. Rev. Lett.* **108**, 074504 (2012).
- [15] R. Lifshitz and H. Diamant, *Philos. Mag.* **87**, 3021 (2007).
- [16] M. Engel and H. R. Trebin, *Phys. Rev. Lett.* **98**, 225505 (2007).
- [17] K. Barkan, H. Diamant, and R. Lifshitz, *Phys. Rev. B* **83**, 172201 (2011).
- [18] C. N. Likos, *Phys. Reports* **348**, 267 (2001).
- [19] J. Dautenhahn and C. K. Hall, *Macromolecules* **27**, 5399 (1994).
- [20] C. N. Likos, H. Löwen, M. Watzlawek, B. Abbas, O. Jucknischke, J. Allgaier, and D. Richter, *Phys. Rev. Lett.* **80**, 4450 (1998).
- [21] A. A. Louis, P. G. Bolhuis, J.-P. Hansen, and E. J. Meijer, *Phys. Rev. Lett.* **85**, 2522 (2000).
- [22] P. G. Bolhuis, A. A. Louis, J.-P. Hansen, and E. J. Meijer, *J. Chem. Phys.* **114**, 4296 (2001).
- [23] A. Jusufi, J. Dzubiella, C. N. Likos, C. von Ferber, and H. Löwen, *J. Phys.: Condens. Matter* **13**, 6177 (2001).
- [24] A. A. Louis, P. G. Bolhuis, R. Finken, V. Krakoviack, E. J. Meijer, and J. P. Hansen, *Physica A* **306**, 251 (2002).
- [25] C. N. Likos and H. M. Harreis, *Condens. Matter Phys.* **5**, 173 (2002).
- [26] I. O. Gotze, H. M. Harreis, and C. N. Likos, *J. Chem. Phys.* **120**, 7761 (2004).
- [27] C. N. Likos, *Soft Matter* **2**, 478 (2006).
- [28] D. A. Lenz, R. Blaak, C. N. Likos, and B. M. Mladek, *Phys. Rev. Lett.* **109**, 228301 (2012).
- [29] A. Lang, C. N. Likos, M. Watzlawek, and H. Löwen, *J. Phys.: Condens. Matter* **12**, 5087 (2000).
- [30] A. J. Archer, C. N. Likos, and R. Evans, *J. Phys.: Condens. Matter* **16**, L297 (2004).
- [31] I. O. Gotze, A. J. Archer, and C. N. Likos, *J. Chem. Phys.* **124**, 084901 (2006).
- [32] B. M. Mladek, D. Gottwald, G. Kahl, M. Neumann, and C. N. Likos, *Phys. Rev. Lett.* **96**, 045701 (2006).
- [33] B. M. Mladek, D. Gottwald, G. Kahl, M. Neumann, and C. N. Likos, *J. Phys. Chem. B* **111**, 12799 (2007).
- [34] A. J. Moreno and C. N. Likos, *Phys. Rev. Lett.* **99**, 107801 (2007).
- [35] C. N. Likos, B. M. Mladek, D. Gottwald, and G. Kahl, *J. Chem. Phys.* **126**, 224502 (2007).
- [36] B. M. Mladek, P. Charbonneau, C. N. Likos, D. Frenkel, and G. Kahl, *J. Phys.: Condens. Matter* **20**, 494245 (2008).
- [37] C. N. Likos, B. M. Mladek, A. J. Moreno, D. Gottwald, and G. Kahl, *Comp. Phys. Com.* **179**, 71 (2008).
- [38] S. D. Overduin and C. N. Likos, *J. Chem. Phys.* **131**, 034902 (2009).
- [39] S. D. Overduin and C. N. Likos, *Europhys. Lett.* **85**, 26003 (2009).
- [40] S. van Teeffelen, A. J. Moreno, and C. N. Likos, *Soft Matter* **5**, 1024 (2009).
- [41] M. Camargo, A. J. Moreno, and C. N. Likos, *J. Stat. Mech.: Theor. and Exp.* **2010**, P10015 (2010).
- [42] M. Camargo and C. N. Likos, *Mol. Phys.* **109**, 1121 (2011).
- [43] A. Nikoubashman, G. Kahl, and C. N. Likos, *Soft Matter* **8**, 4121 (2012).
- [44] R. Evans, *Adv. Phys.* **28**, 143 (1979).
- [45] R. Evans, *Fundamentals of Inhomogeneous Fluids* (Dekker, New York, 1992).
- [46] J. F. Lutsko, *Adv. Chem. Phys.* **144**, 1 (2010).
- [47] U. Marini, B. Marconi, and P. Tarazona, *J. Chem. Phys.* **110**, 8032 (1999).
- [48] U. Marini, B. Marconi, and P. Tarazona, *J. Phys.: Condens. Matter* **12**, A413 (2000).
- [49] A. J. Archer and R. Evans, *J. Chem. Phys.* **121**, 4246 (2004).
- [50] A. J. Archer and M. Rauscher, *J. Phys. A: Math. Gen.* **37**, 9325 (2004).
- [51] R. Evans and M. T. D. Gama, *Molec. Phys.* **38**, 687 (1979).
- [52] A. J. Archer, M. J. Robbins, U. Thiele, and E. Knobloch, *Phys. Rev. E* **86**, 031603 (2012).
- [53] J.-P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic, London, 1986), 2nd ed.
- [54] K. R. Elder, M. Katakowski, M. Haataja, and M. Grant, *Phys. Rev. Lett.* **88**, 245701 (2002).
- [55] Z. F. Huang, K. R. Elder, and N. Provatas, *Phys. Rev. E* **82**, 021605 (2010).
- [56] S. van Teeffelen, R. Backofen, A. Voigt, and H. Löwen, *Phys. Rev. E* **79**, 051404 (2009).
- [57] H. Emmerich, H. Löwen, R. Wittkowski, T. Gruhn, G. I. Tóth, G. Tegze, and L. Gránásky, *Adv. Phys.* **61**, 665

(2012).