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Large two-dimensional clusters of equal-area bubbles: the influence of the boundary in determining the minimum-energy configuration

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ABSTRACT

Finite monodisperse two-dimensional clusters of bubbles are shown to behave like a crystal rather than a liquid. They attain their lowest-energy configuration when they find an arrangement close to a hexagonal lattice, as predicted by the Wulff construction. The deviation from hexagonal shape of the bubbles in a cluster and the variation in bubble pressure throughout the structure are calculated. It is shown that the effect of reducing the length of the boundary is negligible, explaining why configurations retaining the structure of the lattice will give the lowest energy.

§ 1. INTRODUCTION

1.1. *Motivation*

Given many two-dimensional (2D) bubbles of the same area, what is the arrangement that minimizes their total perimeter? For infinitely many bubbles, or with periodic boundary conditions, Hales (2001) showed that it is the honeycomb pattern which minimizes the perimeter. However, for a finite cluster of N bubbles, which is the case of interest for physical 2D foams (in which the energy is proportional to the total line length), the problem is still open. The main difficulty is to find a compromise between the bulk bubbles, which tend to arrange in a honeycomb pattern, and the bubbles at the cluster boundary.

The task is to explore the space of likely candidate clusters and for each of them to measure the line length. For small N, this was performed experimentally up to $N = 15$ (Alfaro *et al.* 1990, Morgan 1994) and then up to $N = 22$ by Vaz and Fortes (2001). Cluster sizes up to $N = 42$ (and also $N = 50$ and 100) were investigated numerically by Cox *et al.* (2003); the method was to cut from a hexagonal lattice

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a finite cluster of N bubbles and then to shuffle it locally by applying a perturbation and performing neighbour-swapping transformations, relaxing at each step.

For larger values of N, we do not know what the actual minimizers are. For $N=100$, the minimal cluster found by Cox *et al.* (2003) presents a honeycomb structure and its outer boundary adopts a sixfold symmetry too. For $N=200$ their shuffling procedure, which could explore only a small fraction of the possible configurations, found a candidate with a circular shape, that is a shorter outer boundary but a more disordered internal structure.

This spurred a debate at the Cambridge 2002 Isaac Newton Institute on Foams and Minimal Surfaces. Do those clusters which retain the (crystal) symmetry of the underlying hexagonal lattice and have a sixfold symmetric boundary (figure 1 (*a*)) have a lower energy than those more disordered (liquid-like) clusters in which the length of the cluster's outer boundary is minimized (figure 1 (*c*))? Is there a critical value of N above which the minimizer's boundary shape switches from sixfold symmetry to circularity?

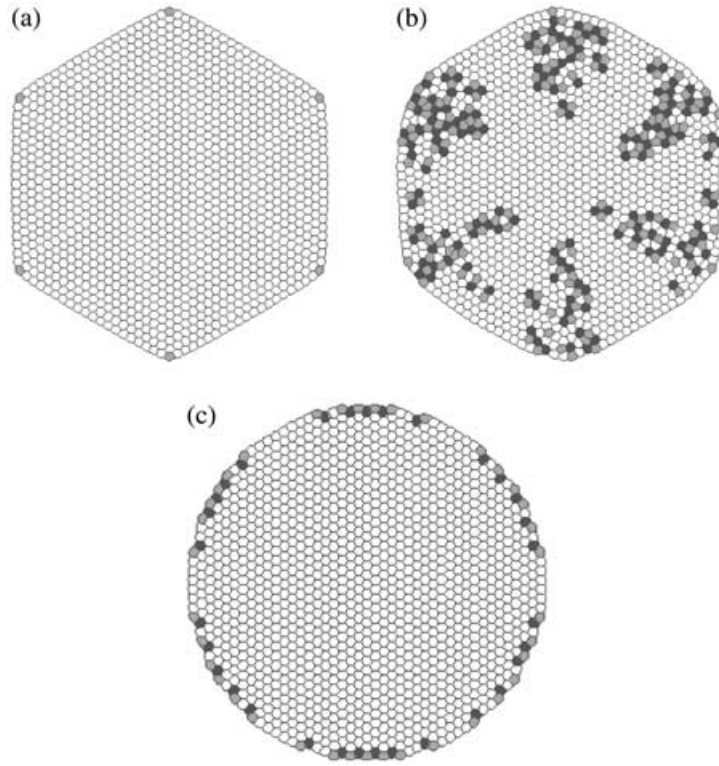

Figure 1. The three methods of preparing clusters, in this case for $N=1261$. In each image, the topological defects are shaded: dark grey for bubbles with a negative topological charge and light grey for those with a positive topological charge. (*a*) The perfect cluster with exactly six four-sided defects. (*b*) A δ-circularized cluster, in this case with $\delta=0.723$. This value of δ creates a partly circularized cluster, with six segments of internal defects. Values of δ of the order of 2.5 would be required to eliminate the ordered rows of peripheral bubbles. (*c*) A circular cluster cut from a hexagonal lattice with many peripheral defects.

Such a transition would not be surprising. Examples exist in related problems, for instance in packings of discs (Schürmann 2002). On the other hand, there is a theoretical argument that suggests the sixfold symmetry remains optimal at arbitrary large N. Fortes and Rosa (2001) calculated the cost of the unit length of a honeycomb's outer boundary and found that it is anisotropic, favouring directions parallel to the honeycomb's close-packed rows. Hence, the Wulff (1901) construction, (Taylor 1994) would predict that the honeycomb lattice should impose its sixfold symmetry and pin the outer boundary.

To find equilibrated clusters and determine their perimeter (or energy), we use the freely available surface-minimizing software, the Surface Evolver (Brakke 1992), in an efficient mode (version 2.18b), adapted to 2D work, which uses arcs of circles to represent each bubble edge. Thanks to this improved version, in the present work we extend the numerical study to large clusters with over 10 000 bubbles and ask what the influence of the periphery of the cluster is in determining its optimal topology. We shall show that a 2D bubble cluster does indeed behave like a crystal rather than a liquid, achieving its minimum-energy configuration when its outer boundary respects the underlying hexagonal lattice rather than assuming a circular shape.

1.2. *Definitions and objectives*

When the topology of the cluster completely respects both the underlying hexagonal lattice and the sixfold symmetry of the boundary, then we refer to the cluster as *perfect*. Such clusters have a regular hexagonal structure consisting of i_0 concentric shells (figure 1 (*a*)) and hence a number of bubbles (Vaz and Fortes 2001)

$$N = 3i_0^2 + 3i_0 + 1. \tag{1}$$

We call these values of N *hexagonal numbers*.

We shall look at perfect clusters, and also at perfect clusters from which one bubble has been removed. We shall then perturb these clusters, making the boundary more circular, to determine the trend of energy change that this causes. This will be compared with the energy of a circular cluster cut from an infinite hexagonal lattice, the starting point for earlier work (Cox *et al*. 2003).

To classify these 2D finite tilings, we describe them in terms of the defects from the hexagonal lattice. We use the notion of topological charge q^*, defined for an s-sided bubble, as introduced by Graner *et al*. (2001). Bubbles in the bulk of the cluster with more than six sides have negative charge, $q^* = 6 - s < 0$; those with less than six sides have positive charge, $q^* > 0$. For bubbles on the periphery, the zero-charge bubbles are those with five sides (i.e. four neighbours plus the external boundary), $q^* = 5 - s$. In each cluster, the total charge is $\Sigma q^* = 6$ (Smith 1952, Graner *et al*. 2001).

For the same hexagonal number N of bubbles, we can prepare different clusters. Examples are shown in figure 1, for $N = 1261$, of

(i) a perfect cluster, with a sixfold symmetric boundary,
(ii) a cluster that has been perturbed to increase the circularity of its boundary, intermediate between (i) and (iii), which may contain internal defects, and
(iii) a circular cluster, cut from a hexagonal lattice.

Below, we shall explain our methods of cluster preparation and then compare their energies.

§ 2. Clusters with a hexagonal number of bubbles

2.1. *Effect of orientation*

We first check that the lowest energy is found when the cluster is aligned with close-packed rows that respect the underlying hexagonal lattice. To do this we start from the infinite lattice and choose a centre point around which we cut $i_0 = 15$ shells ($N = 721$ bubbles) in a hexagonal shape at some angle θ to the lattice; an example is shown in figure 2(*a*). Only values of θ between zero and $\pi/6$ are necessary, by symmetry. For each value of θ we repeat this cut with a different centre point, then relax each of the clusters in the Surface Evolver and record their energies. As shown in figure 2 (*b*), the energy increases rapidly with increasing θ; so the minimal configuration will be found when θ is a multiple of $\pi/3$ and the cluster is perfectly aligned with the underlying lattice. Further, when θ is a multiple of $\pi/3$, there is a cusp in the energy; this is the necessary and sufficient condition to have a facet, as predicted by the Wulff construction (Fortes and Rosa 2001).

2.2. *Perfect clusters*

2.2.1. *Preparation of perfect clusters*

We use a Voronoi construction based on bubble centres with (x, y) coordinates

$$x = 3^{1/2} Li \sin\left(\frac{j\pi}{3}\right) + 3^{1/2} Lk \sin\left(\frac{(j+2)\pi}{3}\right),$$

$$y = 3^{1/2} Li \cos\left(\frac{j\pi}{3}\right) + 3^{1/2} Lk \cos\left(\frac{(j+2)\pi}{3}\right),$$

where i is the shell number between 0 and i_0, j is the number of the side of the hexagon labelled from 1 to 6, and k is the number of the bubble within each of

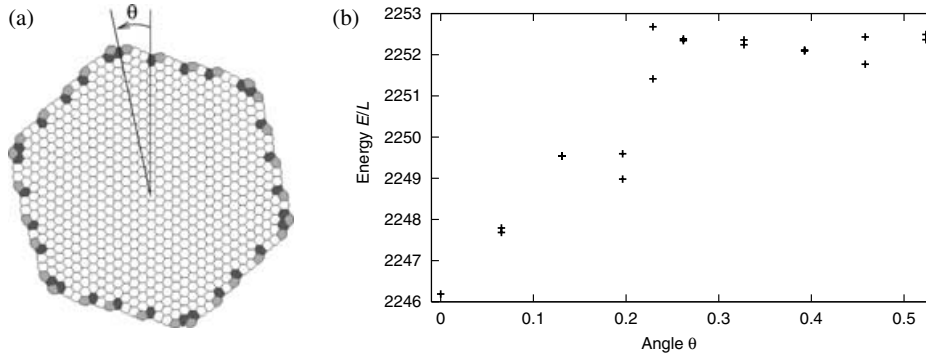

Figure 2. The effect of orientation on a cluster with a hexagonal number of bubbles ascertained by cutting a hexagonal cluster of $N = 721$ bubbles from a hexagonal lattice at different angles θ to the lattice, for $0 \leqslant \theta \leqslant \pi/6$. Figure 1 (*a*) shows the case when $\theta = 0$. (*a*) A hexagonal cluster with $\theta = \pi/12$, showing many surface defects. (*b*) The hexagonal cut is performed twice, with different centre points, for ten values of θ and the energy recorded. It increases rapidly from the value for $\theta = 0$, showing that the minimum-energy configuration should be found when $\theta = 0$. Note the cusp at $\theta = 0$.

the j sides with k varying from 0 to $i-1$. L is the length of one side of a hexagonal bubble of area $A = 3^{3/2}L^2/2$; we choose $L = 1$.

We then create the bubbles around these centres using a Voronoi construction in Mathematica (Wolfram Research 1999); for this purpose we add a small random number (about 0.1) to both x and y to remove the singularity in the Voronoi construction.

2.2.2. *Energy of perfect clusters*

We relax such Voronoi constructions in the Surface Evolver for many hexagonal values of N, constraining each bubble to have area A. We set the surface tension equal to one, so that the energy of a cluster is equal to its line length. We denote the energy of a relaxed perfect cluster of N bubbles by $E_0(N)$, and we expect it to have the lowest possible energy of all such hexagonal-N bubble clusters. In figure 3 we show values of $E - 3N$ from the simulations. We find that the energy of these clusters is well approximated by the honeycomb value $E_h = 3N$, plus a correction due to the boundary:

$$E = 3N + 2\varepsilon(12N - 3)^{1/2}, \tag{3}$$

where $\varepsilon = 0.447$, in excellent agreement with the results of Cox *et al.* (2003), who found $\varepsilon = 0.446 \pm 0.001$, and with those of Vaz and Fortes (2001).

We also find the degree of deviation of the bulk bubbles from regular hexagons in these supposedly optimal clusters. We calculate $\Delta_b = 2E_b - 6N_b$ for several values of N, where E_b is the total edge length of the N_b bubbles in the bulk. As shown in figure 4, we find that Δ_b is always small, $\Delta_b \ll 6N_b$ for $i_0 \leqslant 60$. The relative distortion $2E_b/6N_b - 1$ is less than about 0.1% (see the inset to figure 4).

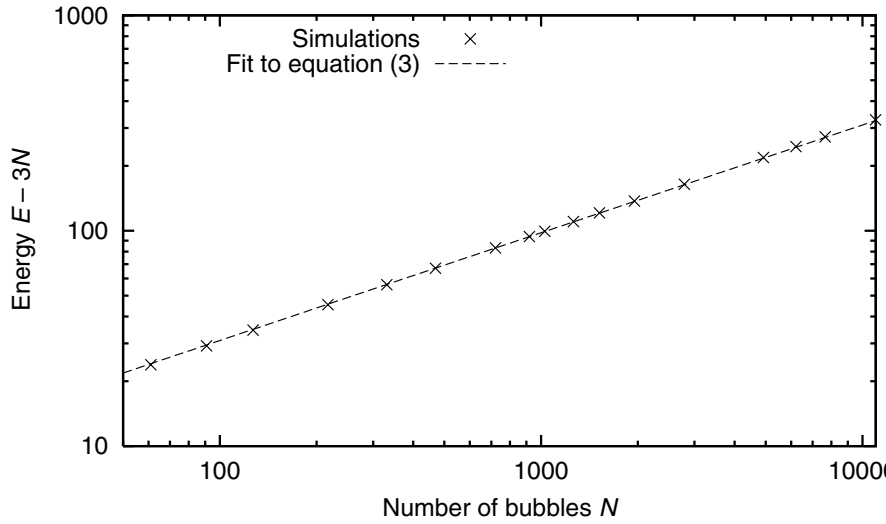

Figure 3. The energy of a perfect cluster versus its number N of bubbles, where N is a 'hexagonal number' given by equation (1), on logarithmic scales. A fit of $\log(E - 3N)$ versus $\log(12N - 3)^{1/2}$, with one adjustable parameter ε, yields $\varepsilon = 0.447$. We thus plot equation (3) with this value (- - - -).

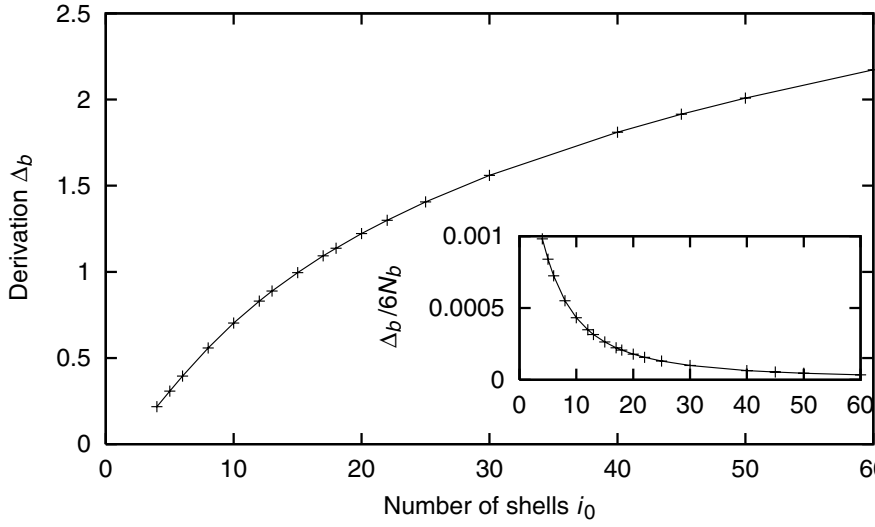

Figure 4. The deviation of the bulk bubbles in a perfect cluster from a hexagonal lattice, measured through the parameter $\Delta_b = 2E_b - 6N_b$. The value of Δ_b increases slowly with increasing number i_0 of shells and is always small compared with the number N_b of bubbles in the bulk, as shown in the inset, so that the bulk bubbles are always close to a hexagonal lattice. The solid curve is a fit of the data by a power law; we find an exponent $i_0^{0.25}$.

Thus, the influence of the boundary in perturbing the bulk hexagons is small, and the centre of a large cluster is close to a hexagonal lattice.

2.2.3. *Bubble pressures in perfect clusters*

The Surface Evolver also allows us to find the pressure in each of the bubbles (compared with an external pressure of zero). We take a large perfect cluster and measure the average pressure, and its standard deviation, in each shell. Performing the same procedure for several hexagonal values of N (i.e. different values of i_0), we find that the effect of the boundary penetrates about one tenth of the way (i.e. around $i_0/10$) into the bulk. For instance, with $i_0 = 60$ shells, figure 5 shows that the pressure is almost constant in the bulk, until approximately 5.6 shells from the boundary. As in § 2.2.2, this suggests that the centre of a large cluster is close to a regular hexagonal lattice.

2.3. *Circularized clusters*

To perform the perturbation (ii), we start again with the Voronoi construct of a perfect cluster. To increase the degree of circularity, we move the boundary from a hexagon to a circle. Then, we displace all bubble centres in proportion to their radial distance from the centre of the cluster. Thus, a Voronoi site at position (x, y) in the perfect hexagonal configuration given by equation (2) moves to

$$x' = r(\theta)\cos\theta, \quad y' = r(\theta)\sin\theta, \tag{4}$$

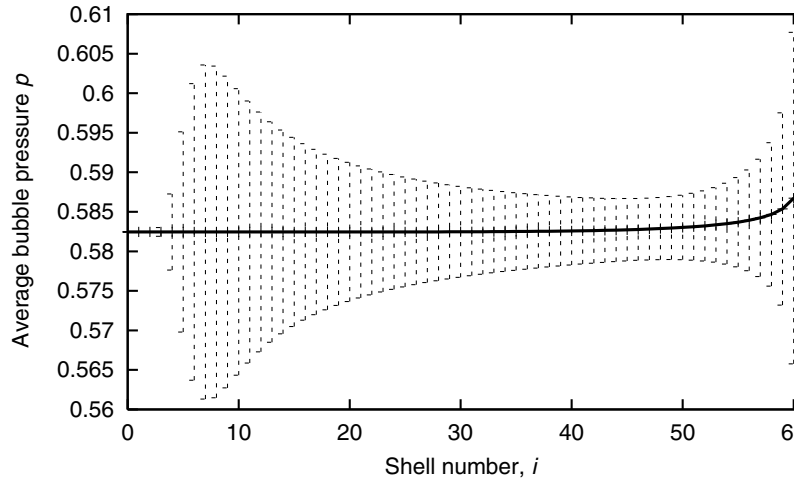

Figure 5. The average pressure per shell (——) for a cluster of $N = 10\,981$ bubbles ($i_0 = 60$ shells). The average pressure increases almost exponentially: $\log(p - p_0) = -16.311 + 0.179\,12i$, for $p_0 = p(i = 0) = 0.582\,45$, suggesting that the effect of the boundary penetrates about $(0.179\,12)^{-1} \approx 5.6$ shells inwards. The standard deviation per shell, plotted as error bars, initially increases, since we are averaging over only a few bubbles, and then decreases to a lower value. As the periphery of the cluster is reached, the deviation again increases, reflecting the effect of the boundary.

where $\tan\theta = y/x$ and

$$r(\theta) = \left(\frac{6(x^2 + y^2)}{3^{1/2}\pi}\right)^{1/2} \cos\left(\left[\theta + \frac{\pi}{6}, \frac{\pi}{3}\right] - \frac{\pi}{6}\right), \tag{5}$$

in which the square brackets denote modulo arithmetic.

However, this does not change the topology and, if we relax this new pattern, it returns to the original sixfold pattern. Thus, before relaxing the pattern we perturb it, so as to introduce defects. We perturb each of the coordinates x' and y' with amplitude δ, which is our control parameter, according to the formula

$$\delta\xi\left(\frac{r(\theta)}{3^{1/2}Li_0}\right)^2, \tag{6}$$

where ξ is a random number between -1 and 1 and i_0 is the total number of shells. With $\delta = 0$ we fall back on sixfold symmetry, while for higher values of δ the pattern becomes more circular (figure 1 (*b*)). The perturbation also increases with increasing radial distance from the cluster's centre. As before, we also add a small random number, to remove the singularity in the Voronoi construction at small δ.

Then we create the Voronoi diagram and import it into the Surface Evolver. We do this for several hexagonal values of N, and many values of δ for each N. The relaxed energies for clusters of $N = 1261$ bubbles are shown in figure 6. They increase almost monotonically with increasing δ, suggesting that the rounding of the cluster and the resulting introduction of defects into the hexagonal structure increase the energy.

We also used these data to obtain the length of the outer boundary of each δ-circularized cluster, which, for $N = 1261$, shows a minimum around $\delta = 0.8$

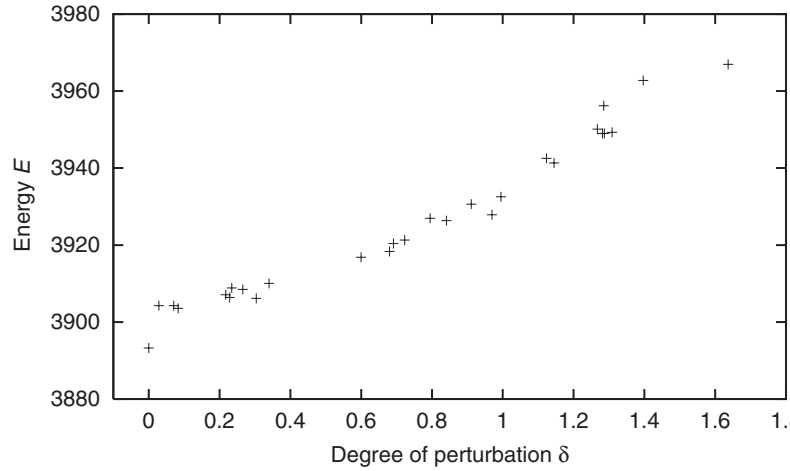

Figure 6. The energy of a δ-circularized cluster of $N = 1261$ bubbles ($i = 20$ shells) for various values of the perturbation parameter δ. The lowest energy occurs for $\delta = 0$, corresponding to the perfect cluster.

(data not shown). So, even when the length of the periphery of the cluster is minimized, the energy of the whole cluster is not. This again suggests to us that the boundary is not so significant in determining the best arrangement of bubbles.

2.4. *Circular clusters*

To make the circular cluster in (iii), we take a large hexagonal lattice of side length $L = 1$ in the Surface Evolver and perform the process described by Cox *et al.* (2003). That is, we take a large circle and slowly decrease its radius, at each step deleting a single bubble as its centre moves outside the circle. There is no unique choice for the position of the centre of the circle, provided that it does not correspond to a centre of symmetry of the lattice. We find that the value of energy for a circular cut is *always* greater than that for a perfect cluster, although the choice of centre can affect quite significantly the final value of energy. For example, with $N = 469$ ($i_0 = 12$) the energy excess over the value for the perfect cluster varies from $E_c(469)$–$E_0(469) = 1.32$ to 2.37. These differences are shown in figure 7 for various values of N and up to three cuts with different centres.

§ 3. Other clusters

3.1. *Perfect clusters less one bubble*

To investigate the best cluster for a case in which there is not a hexagonal number of bubbles, we consider the 20-shell perfect cluster with one bubble removed, that is $N = 1260$ bubbles. Assuming that the best cluster for $N = 1261$ bubbles is the perfect cluster, from where should we remove a bubble to obtain the best configuration for $N = 1260$? We perform a circular cut of 1260 bubbles from a hexagonal lattice, case (iii), three times and record the energy. Then we take the perfect cluster and remove a single bubble from each of several different places, shown in figure 8 (*a*). To remove a bubble, we delete the corresponding bubble centre, create the Voronoi construction and then relax the new configuration. The optimal place to remove the bubble, such that the resulting cluster has the lowest energy, is from a peripheral

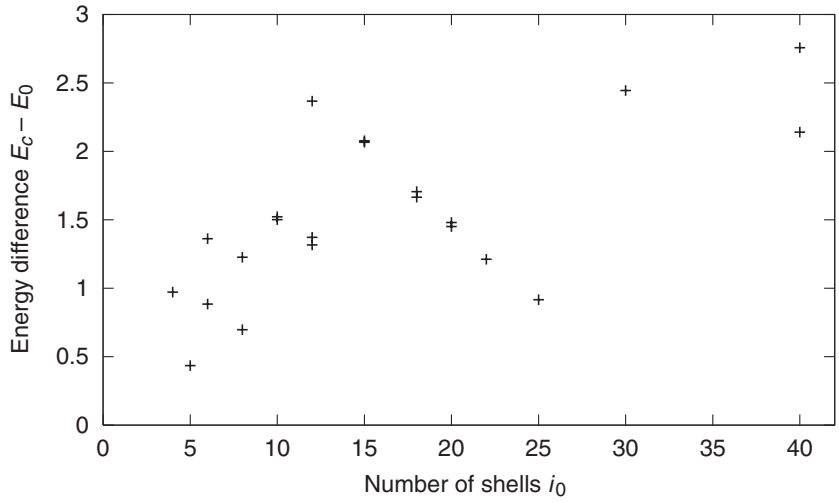

Figure 7. For a perfect cluster consisting of i_0 shells, where the number N of bubbles is hexagonal (equation (1)), we show the difference in energy (always positive) between various circular cuts (figure 1 (*c*)) and the sixfold symmetric arrangement (figure 1 (*a*)).

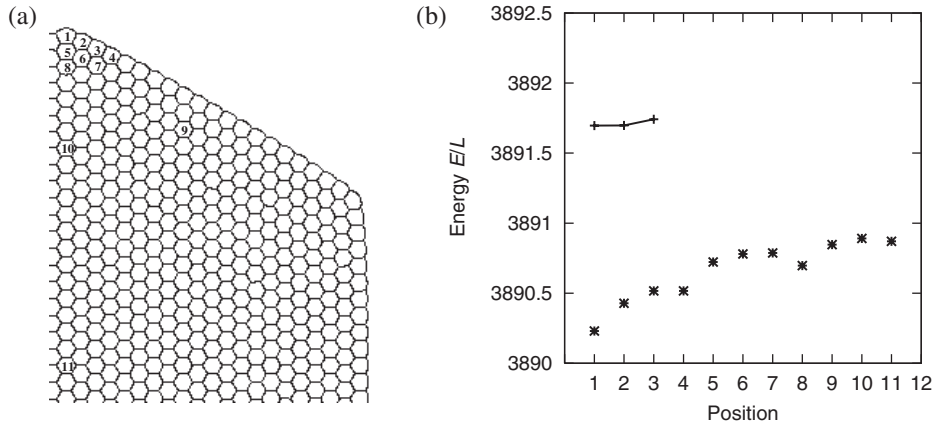

Figure 8. We remove one bubble from a perfect cluster of 1261 bubbles to determine the optimal structure of the cluster for $N = 1260$. (*a*) Bubbles are removed from the positions indicated in the diagram. (*b*) The energy of the relaxed cluster recorded (✱). Also shown is the energy of a circular cluster cut from a hexagonal lattice (+—+), with three choices for the centre of the circle (unrelated to the positions in (*a*)), which is always greater. The best place to remove a bubble is from the apex at one corner (position 1).

corner of the perfect cluster (see figure 8 (*b*)). Removing a single bubble from a perfect cluster always results in a lower energy than the circular cut.

3.2. $N = 200$

We extend the notion of finding the best cluster for non-hexagonal N by looking at the case $N = 200$ (see the introduction) discussed by Cox *et al*. (2003). This value of N lies far from a hexagonal number and hence there is a better chance that the circular cluster has a lower energy.

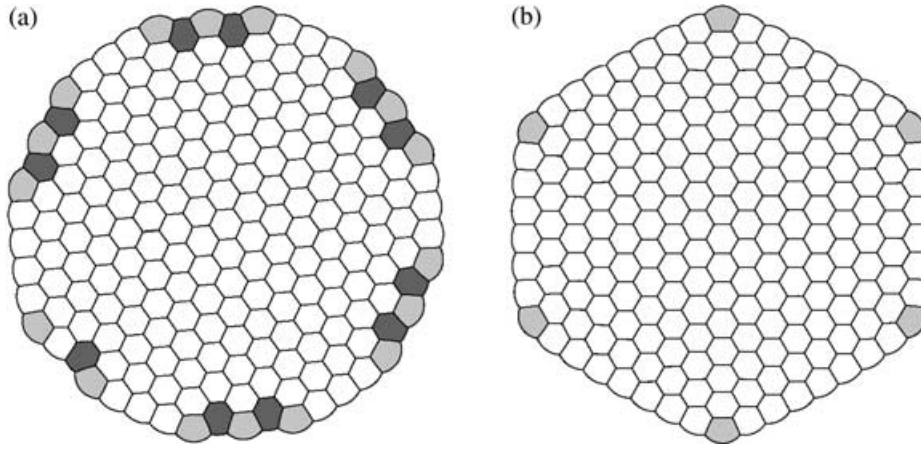

Figure 9. Removal of 17 bubbles from a perfect cluster of 217 bubbles to find the best cluster of $N=200$ bubbles. (*a*) A circular cut from a hexagonal lattice. (*b*) A lower-energy configuration with a honeycomb-like structure and a boundary that is pinned to the hexagonal lattice.

The shuffling procedure described by Cox *et al.* (2003) found a circular cluster with $E=644.179$ (figure 9(*a*)) and 11 topological defects around the periphery. These authors suggested that their shuffling procedure was ineffective for this larger value of N. We therefore used our new methods to look at this case.

We find a better configuration (figure 9(*b*)) which, although it is not sixfold symmetric, does retain the pinning of the boundary onto the underlying hexagonal lattice. It is obtained from a perfect cluster of 217 bubbles by 'shaving' a row of peripheral bubbles off two sides (by removing, as above, the corresponding points before using the Voronoi construction), to give a configuration with a value of energy that is lower by about 0.1%: $E=643.562$. (Note that, even though the latter configuration has fewer peripheral bubbles, the total length of the boundary is still greater.) Moreover, it is consistent with the conjecture of Cox *et al.* (2003) that the best candidate configurations should minimize the number of negative defects.

3.3. $N=1000$

To check our methods and conclusions, we do the same for the case $N=1000$, removing 27 bubbles from the perfect 18-shell cluster. We try ten different configurations and find that the optimum arrangement, shown in figure 10, is to remove bubbles surrounding *two* corners, the total length of one side, and equal parts of two other sides. This has lower energy than, for instance, removing bubbles starting from one corner so that there is only one defect, or a circular cut from a hexagonal lattice. There are two defects in this configuration, so that in this case our candidate minimal cluster might not minimize the number defects.

3.4. $N=1081$

One might ask whether, for values of N far from a hexagonal number, a polygon with a degree of symmetry other than sixfold might offer an arrangement with lower energy. As an example, we cut a 12-sided cluster of $N=1081$ bubbles from the hexagonal lattice. This cluster, shown in figure 11(*a*), has a boundary consisting of alternating arrangements of close-packed rows and regions of surface defects; we

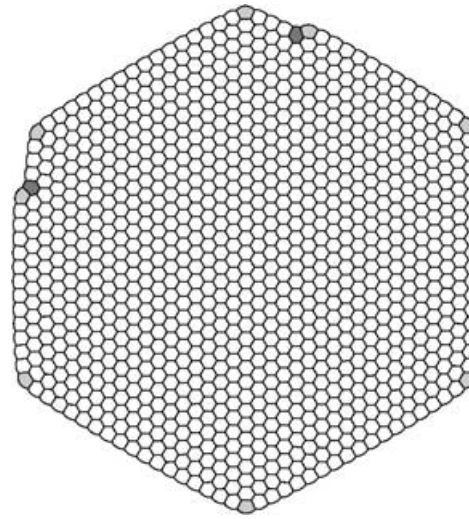

Figure 10. The best candidate configuration found for $N = 1000$. Note that, as well as deleting bubbles from one complete side of the perfect cluster with $N = 1027$, we also take bubbles from two corners and parts of two adjacent sides.

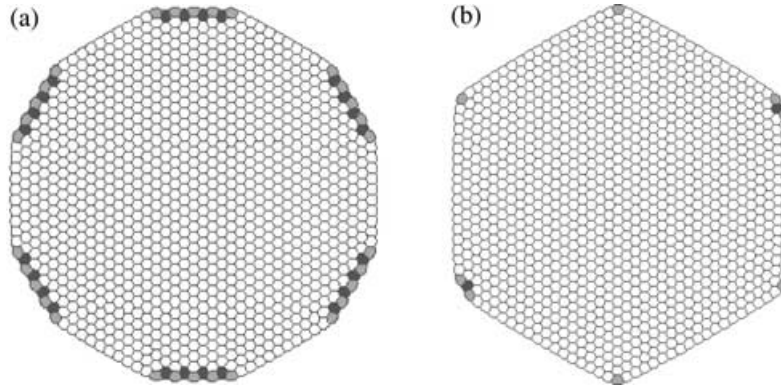

Figure 11. A comparison of (*a*) a 12-sided cluster of $N = 1081$ bubbles with (*b*) the corresponding six-sided cluster. The latter has fewer surface defects and lower energy.

choose it precisely because of the regular surface pattern and because the number of bubbles is far from a hexagonal number. After relaxation in the Surface Evolver, we find an energy of $E = 3345.264$. We compared this with the energy of a perfect 19-shell cluster from which three sides are removed to leave the same number of bubbles, shown in figure 11 (*b*). The latter has a *lower* energy of $E = 3344.962$, again showing the importance of adhering to the hexagonal lattice everywhere and reducing the number of defects.

§4. Conclusions

A finite monodisperse 2D foam (N bubbles with equal areas) behaves like a crystal rather than a liquid. That is, its minimal energy is achieved when its outer boundary respects the underlying hexagonal lattice, rather than by introducing internal 5/7 defect pairs and reducing its boundary energy.

We find that the inside of the cluster must be close to the honeycomb, and that the minimum energy configuration, for $N \leqslant 10981$ (and surely for greater N), is given by the closest possible configuration to a regular honeycomb-like structure. Is it possible that far from a hexagonal number we could find a better configuration than that respecting the honeycomb lattice? The results for $N = 200$, 1000 and 1081, and our intuition, suggest not. We thus confirm the theoretical argument derived by Fortes and Rosa (2001).

What should happen when we relax the condition of monodispersity? We presume that such area dispersity will lead to topological dispersity (non-hexagonal bubbles) and the consequent loss of the correlations in geometrical ordering. We therefore expect a transition to liquid-like behaviour and a circular boundary to the cluster. This is what Fortes and Rosa (2001) suggested according to their calculations of the cost of the outer boundary for polydisperse disordered foams.

In three dimensions, the problem of finding optimal arrangements still remains remarkably difficult since, beside the computational difficulties involved in finding an efficient search algorithm, we do not even know the minimizer for an infinite number of bubbles.

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