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Lower bounds for the surface energy of two-dimensional foams

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Abstract. Amongst the two-dimensional cellular patterns that fill a plane, dry foams at stable equilibrium typify a particular subset for which the total perimeter P of cell boundaries (*i.e.*, films between bubbles) has a local minimum. For a given set of bubble areas A_i ($i = 1, \ldots, N$), P can be written in the form $P = R(\sum_{i=1}^{N} \sqrt{A_i})/2$, where R is topology dependent. We seek the set of areas A_i and the cluster topology that minimise R, and propose lower bounds for R that set lower bounds for the surface energy of i) individual bubbles, with circular edges meeting at $2\pi/3$ angles at vertices (Plateau cells), and ii) infinite periodic bubble clusters.

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1 Introduction

A liquid foam is an aggregate of gas bubbles, or cells, bounded by liquid films. Foams have many important industrial applications, ranging from food, toiletries and cleaning products, to fire fighting, oil recovery and mixture segregation by fractionation or flotation [1]. In addition, foams at (stable) equilibrium are a model for a class of surface-dominated systems that minimise the area (in three dimensions) or the perimeter (in two dimensions) of interfaces. A two-dimensional (2d) foam —a bubble cluster— consists of N bubbles (per unit cell, in the case of a periodic foam) of areas A_i ($i = 1, \ldots, N$). We focus on timescales much shorter than those typical of foam coarsening, so the bubbles preserve their integrity and size. Furthermore, we assume that the bubble gas is incompressible; the area of a bubble is thus independent of its location in the cluster, and the gas energy is constant. The energy of a dry cluster is then just the surface energy,

$$E = P\gamma, \tag{1}$$

where P is the total length of films and γ is the film tension. In an unbounded cluster, $P = \frac{1}{2}\sum_{i=1}^{N} P_i$, where P_i is the perimeter of bubble i, as each film is shared between two bubbles; in a finite cluster, by contrast, the peripheral films are not shared and $P > \frac{1}{2}\sum_{i=1}^{N} P_i$.

Each 2d foam at stable equilibrium realises a tiling of the plane which is a local minimum of the perimeter or surface energy, equation (1). In such a foam, *Plateau's laws* [2] are satisfied: i) films meet three at a time at $2\pi/3$ angles at vertices, and ii) each film between two bubbles, say i and j, is an arc of circle of curvature κ_{ij} determined by the pressure difference $\Delta p = p_i - p_j$ between the bubbles: $\kappa_{ij} = \Delta p/\gamma$. Consequently, the sum of all three curvatures at a vertex is zero. We call a bubble that obeys these laws a *Plateau cell*. Figure 1 shows the different types of Plateau cells considered in this paper: (a) is a *regular* Plateau cell (all sides are identical); (b), (c) and (d) are non-regular, but all are highly symmetric.

We focus particularly on the set of 2d unbounded clusters (or on bounded clusters large enough to contain only a negligible fraction of peripheral bubbles). For a given distribution of bubble areas, different bubble arrangements —*i.e.*, topologies— may be possible, and for each topology the cluster may be strained or unstrained. In general, unstrained clusters will have lower energies. Clusters with the same bubble area distribution but different topologies are composed of bubbles of different shapes (all of which are Plateau cells), and hence generally have different total perimeter.

For a given set of N areas $\{A_i\}$ and given topology, we write, for the total perimeter of an unstrained cluster,

$$P = R\,\frac{\sum_{i=1}^{N} \sqrt{A_i}}{2}\ , \tag{2}$$

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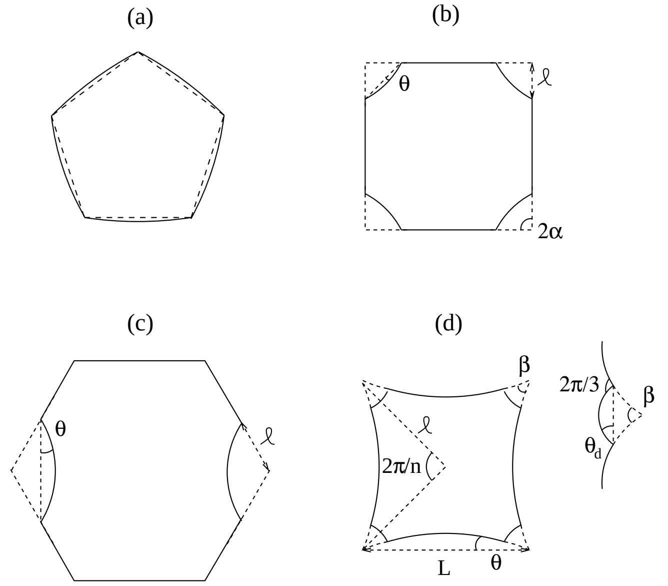

Fig. 1. (a) Regular, 5-sided Plateau cell. (b) Vertex-decorated Plateau cell with straight and curved sides. (c) Regular hexagon decorated at $\nu = 2$ vertices. (d) Vertex-decorated Plateau cell with two families of curved sides ($n = 4$); the inset shows the detail of the decoration —it is such that all internal angles are $2\pi/3$.

where R is a (topology-dependent) number; it is of course independent of scale. The main purpose of this paper is to find a better lower bound for R (which presumably will be realised in an unbounded unstrained cluster). A lower bound for R is known in the case of a monodisperse foam (*i.e.*, $A_i = A$ for all i): this is realised by regular hexagonal cells with straight sides [3], for which $R_{\text{hex}} = P_{\text{hex}}/\sqrt{A_{\text{hex}}} = 2^{1/2}3^{1/4} \approx 3.722419/2$.

For a cluster with an arbitrary distribution of bubble areas, what bounds can we propose? Two are currently available. The first bound is very conservative, but rigorous. The absolute minimum $P/\sqrt{A}$ of a closed curve is that of a circle, $R_{\text{circle}} = 2\sqrt{\pi} \approx 3.544908$. Therefore, any Plateau cell has $R_{\text{PC}} \geq R_{\text{circle}}$, so that for any cluster (bounded or unbounded),

$$P \geq 2\sqrt{\pi}\,\frac{\sum_{i=1}^{N}\sqrt{A_i}}{2} \approx 3.544908\,\frac{\sum_{i=1}^{N}\sqrt{A_i}}{2}. \quad (3)$$

The second bound is an earlier conjecture by one of the present authors [4]. It originated with the calculation of $R(n) \equiv P/\sqrt{A}$ for regular n-sided Plateau cells, which consist of n identical arcs of circle meeting at internal angles of $2\pi/3$. $R(n)$ is a weakly decreasing function of n, with $R(6) = 2R_{\text{hex}} = 2^{3/2}3^{1/4} \approx 3.722419$ and $R(\infty) = 2\pi^{3/2}/3 \approx 3.712219$. This suggested that, for any cluster,

$$P \geq \frac{2\pi^{3/2}}{3}\,\frac{\sum_{i=1}^{N}\sqrt{A_i}}{2} \approx 3.712219\,\frac{\sum_{i=1}^{N}\sqrt{A_i}}{2}. \quad (4)$$

We shall show that the above conjecture is false: unlike straight-sided polygons, Plateau cells are not optimal when regular —there are non-regular Plateau cells with $R \equiv P/\sqrt{A}$ below $2\pi^{3/2}/3$, the smallest R for regular Plateau cells.

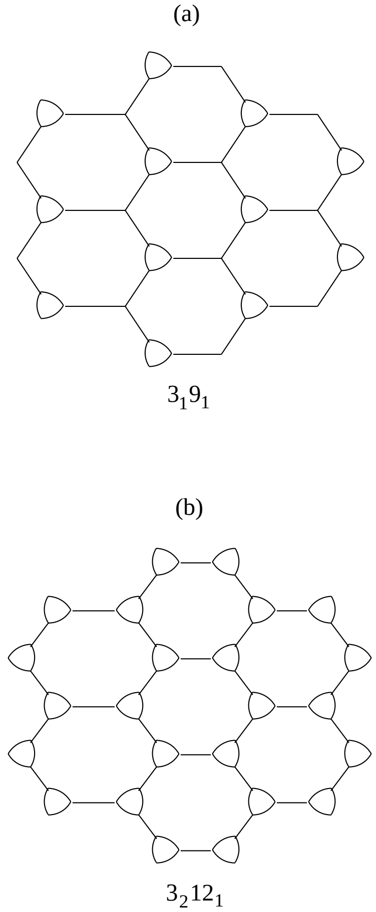

Fig. 2. (a) $3_1 9_1$ and (b) $3_2 12_1$ tilings. The latter realises our conjectured lower bound for $P/(\sum_i \sqrt{A_i})$ of a partition of the plane into regions of two different areas, for area ratio $\lambda = A_3/A_{12} = 0.008195$ (Eq. (29)).

Moreover, a (periodic) cluster with $R < 2\pi^{3/2}/3$ has been found recently [5]: it is composed of one 3-sided cell and one 9-sided cell per period, and is denoted $3_1 9_1$. Its topology is shown in Figure 2a. In [5] we calculated R for this cluster as a function of $\lambda \equiv A_3/A_9$, see Figure 3, and found a range where the lower bound of [4] is violated: the lowest $R = 3.70611$ is attained when $\lambda \approx 0.008821$. We are thus in a position to state that any lower bound R^* on the R of an arbitrary cluster (*i.e.*, of arbitrary area distribution and arbitrary topology) will be such that

$$3.544908 \leq R^* \leq 3.70611. \quad (5)$$

As mentioned above, there exist Plateau cells whose $P/\sqrt{A}$ is less than that of a regular Plateau cell. Here we search for the Plateau cells of the smallest $P/\sqrt{A}$.

This paper is organised as follows: in Section 2 we study isolated Plateau cells of small $R_{\text{PC}} \equiv P/\sqrt{A}$ and show that it is possible to construct such a cell with R_{PC} asymptotically approaching R_{circle}. In Section 3 we discuss

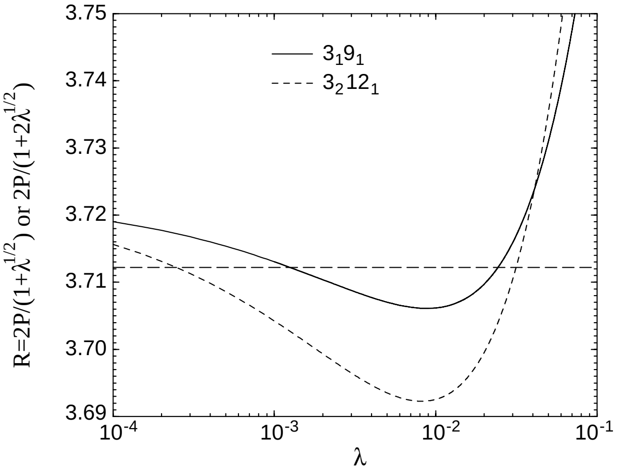

Fig. 3. Ratio R (defined in Eq. (2)) for tilings $3_1 9_1$ (see Fig. 2a) and $3_2 12_1$ (see Fig. 2b), plotted as a function of the cell area ratios $\lambda \equiv A_3/A_9$ [5] or $\lambda \equiv A_3/A_{12}$. Both violate Graner *et al.*'s bound (long-dashed line, Eq. (4)), in the ranges $0.0012 \lesssim \lambda \lesssim 0.0243$ and $0.0002 \lesssim \lambda \lesssim 0.0315$, respectively.

unbounded clusters composed of (space-filling) Plateau cells and present one that has R slightly below that of the $3_1 9_1$ cluster. In Section 4 we summarise our results, which consist of two new lower bounds for the R of i) a single Plateau cell, and ii) an unbounded, unstrained cluster. We carefully distinguish what is rigorous and what is conjectured; note, however, that in this field, where mathematical proofs are notoriously difficult to accomplish, conjectures made on physical grounds can play, and have played, an important role: indeed even such milestones as Plateau's laws [6] and the honeycomb conjecture [3] have received formal confirmation only very recently.

2 Plateau cells of least perimeter

In this section we discuss three types of regular (isolated) Plateau cells as candidates to yielding a lower bound for $R_{\rm PC} = P/\sqrt{A}$: in Section 2.1, regular $2n$-sided cells with n straight sides alternating with n circular sides, constructed by decorating the vertices of an n-sided regular polygon with circular films meeting the straight sides at $2\pi/3$ angles (see Fig. 1b); in Section 2.2, regular hexagonal (straight-sided) cells with ν (≤ 6) vertices decorated with arcs of circle meeting the straight sides at $2\pi/3$ angles (see Fig. 1c); and in Section 2.3, as in Section 2.1, but with the straight sides replaced by arcs of circle meeting at $2\pi/3$ (see Fig. 1d). We shall show that cells of the last type exist, whose $R_{\rm PC}$ approaches the absolute minimum of $P/\sqrt{A}$ for any closed curve (*i.e.*, its value for a circle).

2.1 Straight-sided n-cells with decorated vertices

We start with a regular n-cell with straight sides (an n-sided polygon); the vertices are then "decorated" with arcs of circle that meet the straight sides at $2\pi/3$ (so that the resulting cell is a Plateau cell), as in Figure 1b. If ℓ is the length of a straight side replaced by the decoration, we then have the following equations for the perimeter P and area A of the decorated cell with $2n$ sides, taking a, the side length of the original cell, as our unit of length (*i.e.*, $a = 1$):

$$P = n\left(1 - 2\ell C\right), \tag{6}$$
$$A = n\left(D - E\ell^2\right), \tag{7}$$

where

$$C = 1 - \sin\alpha\,\frac{\theta}{\sin\theta}, \tag{8}$$
$$D = \frac{1}{4}\cot\frac{\pi}{n}, \tag{9}$$
$$E = \sin^2\alpha\,\frac{\theta - \sin\theta\cos\theta}{\sin^2\theta} + \sin\alpha\cos\alpha, \tag{10}$$

and

$$\alpha = \frac{\pi}{2}\left(1 - \frac{2}{n}\right) \tag{11}$$

is half the internal angle of the original n-sided polygon, and

$$\theta = \pi\left(\frac{1}{3} - \frac{1}{n}\right) = \alpha - \frac{\pi}{6} \tag{12}$$

is half the subtended angle of the arcs of circle decorating the vertices. Minimising $R_{\rm PC} = P/\sqrt{A}$, we obtain

$$\ell^* = \frac{2CD}{E}, \tag{13}$$

and the corresponding value of $R_{\rm PC}$ at the minimum, $(R_{\rm PC})_{\rm min}$, is

$$(R_{\rm PC})_{\rm min} = \sqrt{\frac{n}{D}\left(1 - \frac{4C^2 D}{E}\right)}. \tag{14}$$

$(R_{\rm PC})_{\rm min}$ are collected in the fourth column of Table 1 for $3 \leq n \leq 8$; the lowest minimum, $(R_{\rm PC})_{\rm min} = 3.640455$, is attained for $n = 8$. If $n \geq 9$, $C < 0$ and $R_{\rm PC}$ is minimised for $\ell = 0$, *i.e.*, in the straight-sided n-cell.

2.2 Decorated hexagonal cells

Let us now consider Plateau cells based on the regular hexagon (*i.e.*, with $2\pi/3$ internal angles), constructed by decorating ν of its vertices ($0 \leq \nu \leq 6$) with circular films. Each of these replaces a length ℓ of the original straight sides, with which it makes $2\pi/3$ angles (see Fig. 1c); the resulting Plateau cells have $6 + \nu$ sides. These fill space when combined with 3-cells, see below.

Taking the side length of the original hexagon as unity, we obtain

$$\frac{P^2}{A} = \frac{2}{3\sqrt{3}}\left[\frac{(6 - \nu\lambda_1\ell)^2}{1 - \nu\lambda_2\ell^2}\right], \tag{15}$$

with

$$\lambda_1 = 2 - \frac{\pi}{\sqrt{3}}\ ; \quad \lambda_2 = \frac{\pi - \sqrt{3}}{3\sqrt{3}}. \tag{16}$$

Table 1. Minima of $R_{\rm PC} = P/\sqrt{A}$ for the different single Plateau cells considered, compared with $P/\sqrt{A}$ for regular straight-sided polygons. n in column 1 is the total number of sides of the actual cell: in column 4 this is twice the number of sides of the original, undecorated, polygon, whereas in column 5 it is the number of sides of the decorated hexagon (*i.e.*, $n = \nu + 6$, where ν is the number of decorated vertices). See the text for details.

n	Regular n-polygon (straight sides)	Plateau cells: Regular (Fig. 1a)	Plateau cells: Decorated $(n/2)$-polygon (Fig. 1b)	Plateau cells: Hexagon decorated at $\nu = n - 6$ vertices (Fig. 1c)
3	4.559014	3.742190	–	–
4	4.000000	3.730802	–	–
5	3.811935	3.725463	–	–
6	3.722419	3.722419	3.722419	3.722419
7	3.672069	3.720473	–	3.715806
8	3.640719	3.719130	3.714018	3.709180
9	3.619797	3.718151	–	3.702543
10	3.605106	3.717409	3.700116	3.695894
11	3.594380	3.716827	–	3.689232
12	3.586302	3.716360	3.682559	3.682559
13	3.580063	3.715977	–	–
14	3.575141	3.715658	3.662448	–
15	3.571189	3.715388	–	–
16	3.567967	3.715156	3.640455	–
∞	$2\pi^{1/2} \approx 3.544908$	$\frac{2}{3}\pi^{3/2} \approx 3.712219$	–	–

The minimum of P^2/A is for $\ell = \ell^*$, independent of ν:

$$\ell^* = \frac{\lambda_1}{6\lambda_2} \approx 0.114402. \tag{17}$$

$(R_{\rm PC})_{\rm min} \equiv P/\sqrt{A}$ at the minimum are given in the fifth column of Table 1. $(R_{\rm PC})_{\rm min}$ decreases with increasing ν, attaining its smallest value $(R_{\rm PC})_{\rm min} = 3.682559$ for $\nu = 6$.

2.3 Curved-sided n-cells with decorated vertices

Finally, consider a regular cell with n circular sides, each of radius r and subtended angle 2θ, and such that two adjacent sides meet at an internal angle

$$\beta = \pi\left(1 - \frac{2}{n}\right) - 2\theta, \tag{18}$$

which need not equal $2\pi/3$. This is illustrated in Figure 1d; θ is defined as positive if the centres of curvature of the sides are outside the cell. We next construct a Plateau cell by decorating all vertices with arcs of circle of radius $r_{\rm d}$ and subtended angle $2\theta_{\rm d}$ to obtain $2\pi/3$ angles at every (new) vertex (see inset of Fig. 1d), which requires

$$\theta_{\rm d} = \frac{\beta}{2} - \frac{\pi}{6}. \tag{19}$$

It will now be shown that there are Plateau cells of this type whose $R_{\rm PC} = P/\sqrt{A}$ approaches the absolute minimum $R_{\rm circle} = 2\sqrt{\pi} \approx 3.544908$. We can then conclude that the lower bound for $R_{\rm PC}$ coincides with the absolute minimum. It is attained in the limit $r_{\rm d} \to 0$, $n \to \infty$ and $\theta \to 0$, for which $\theta_{\rm d} \to \pi/3$. To see this, start by noting that, if $r_{\rm d} \sim 0$, then decorated and undecorated cells have the same perimeter and the same area:

$$P = 2nr\theta, \tag{20}$$

$$A = nr^2\left[\sin^2\theta\,\cot\frac{\pi}{n} - (\theta - \sin\theta\,\cos\theta)\right], \tag{21}$$

where we have used the relation (see Fig. 1d for the definition of ℓ and L)

$$\frac{L}{2} = \ell\sin\frac{\pi}{n} = r\sin\theta. \tag{22}$$

Equations (20) and (21) lead to

$$\frac{P}{\sqrt{A}} = 2\theta\sqrt{\frac{n\tan\frac{\pi}{n}}{\sin^2\theta - \tan\frac{\pi}{n}\,(\theta - \sin\theta\,\cos\theta)}}. \tag{23}$$

As $n \to \infty$, $\tan\frac{\pi}{n} \to 0$ and $n\tan\frac{\pi}{n} \to \pi$; and as $\theta \to 0$, $\theta/\sin\theta \to 1$, whence

$$R^*_{\rm PC} = \lim_{n\to\infty}\frac{P}{\sqrt{A}} = 2\sqrt{\pi}. \tag{24}$$

This is the $P/\sqrt{A}$ of a circle and is therefore a firm lower bound for the perimeter of a Plateau cell. Hence for any Plateau cell,

$$\frac{P}{\sqrt{A}} \geq 2\sqrt{\pi}, \tag{25}$$

which bound cannot be improved.

3 Clusters of least perimeter

The foregoing analysis provides a guide to finding a lower bound R^* for the R of a cluster, and thus for its perimeter (or energy) for arbitrary bubble areas and arbitrary topology. Here we concentrate on unbounded clusters because their perimeter is generally smaller: in finite clusters the peripheral films are not shared between two cells, which, as we have already noted, penalises R [4] (in other words, an outer boundary costs energy).

In the preceding section we identified individual Plateau cells of very small $R_{\rm PC}$, see Table 1, for which we found the lower bound $R^*_{\rm PC} = 2\sqrt{\pi}$. But these have to be combined with cells of larger $R_{\rm PC}$ in order to fill space. Better candidates for the lower bound are thus periodic tilings based on the regular honeycomb, with truncated vertices. This is suggested by our finding [5] that the $3_1 9_1$ tiling has particularly low R, providing the first known violation of the lower bound expressed in equation (4). So it is to be expected that the smallest R will be achieved when all vertices of a honeycomb are decorated, leading to a $3_2 12_1$ tiling as pictured in Figure 2b. We have calculated R for tilings $3_1 9_1$ and $3_2 12_1$.

We then have, for the $3_1 9_1$ tiling,

$$R = \frac{2P}{\sqrt{A_3} + \sqrt{A_9}} = \frac{2\left[3 + \sqrt{3}\left(\pi - \sqrt{3}\right)\ell\right]}{\ell\sqrt{\frac{3}{2}\left(\pi - \sqrt{3}\right)} + \sqrt{\frac{3}{2}\left[\sqrt{3} - \left(\pi - \sqrt{3}\right)\ell^2\right]}}, \qquad (26)$$

and for the $3_2 12_1$ tiling,

$$R = \frac{2P}{2\sqrt{A_3} + \sqrt{A_{12}}} = \frac{2\left[3 + 2\sqrt{3}\left(\pi - \sqrt{3}\right)\ell\right]}{2\ell\sqrt{\frac{3}{2}\left(\pi - \sqrt{3}\right)} + \sqrt{\frac{3}{2}\left[\sqrt{3} - 2\left(\pi - \sqrt{3}\right)\ell^2\right]}}, \qquad (27)$$

where we have taken as unit of length the side of the original hexagons, and ℓ is as in Figure 1c. Plots of R *vs.* the cell area ratios λ as defined below, are shown in Figure 3. Minimisation yields, for the $3_1 9_1$ tiling,

$$\ell_{\rm min} = 0.103657\,;\ \lambda \equiv \frac{A_3}{A_9} = 0.008821\,;\ R_{\rm min} = 3.70611, \qquad (28)$$

which confirms the result of Fortes and Teixeira [5]; and, for the $3_2 12_1$ tiling,

$$\ell_{\rm min} = 0.0995398\,;\ \lambda \equiv \frac{A_3}{A_{12}} = 0.008195\,;\ R_{\rm min} = 3.69228. \qquad (29)$$

As anticipated, R is smallest when all vertices are decorated.

We have also checked whether triangle decoration of fewer vertices in the honeycomb could lower this bound. Consider a $n_1 \times n_2$ subarray of the honeycomb containing $n_1 n_2$ cells and $2n_1 n_2$ vertices. Decorate $2n_1 n_2 - 1$ vertices with regular triangular Plateau cells, all of the same area; there are now also three 11-sided cells in the motif. For any choice of n_1 and n_2, we found R to be greater than that for the $3_2 12_1$ tiling. We expect R to increase if fewer than $2n_1 n_2 - 1$ vertices are decorated; the smallest R should be attained when all vertices of the honeycomb are decorated with triangles, *i.e.*, in the $3_2 12_1$ tiling.

None of the Plateau cells discussed in Sections 2.1-2.3 which have $R_{\rm PC}$ lower than that of the $3_2 12_1$ tiling is space filling. We therefore conjecture that the $R_{\rm min}$ of the $3_2 12_1$ tiling with $\lambda = A_3/A_{12} = 0.008195$ is the true lower bound for the R of a bubble cluster:

$$R^* = 3.69228, \qquad (30)$$

which is 0.5% lower than Graner *et al.*'s [4]. The new, conjectured lower bound on the surface energy of a foam cluster is then

$$E = P\gamma \geq 3.69228\,\frac{\sum_{i=1}^{N}\sqrt{A_i}}{2}\,\gamma. \qquad (31)$$

4 Conclusions

Stable equilibrium foam structures are the result of surface energy minimisation. Now that it has been proved that the regular honeycomb is optimal for monodisperse 2d foams [3], it is natural to look at the polydisperse case. We have identified a number of instances where the lower bound conjectured by Graner *et al.* for the ratio R (defined by Eq. (2)) of a 2d dry foam is violated. On the basis of our analysis we propose new lower bounds for the R (hence for the surface energy) of i) a single Plateau cell, and ii) a dry, bidisperse 2d foam of arbitrary bubble areas. Bound i), equation (25), is realised in a Plateau cell whose boundaries consist of infinitely many arcs of two distinct families, with the common radius of the arcs belonging to one of these families going to zero. This coincides with the absolute minimum for the $P/\sqrt{A}$ ratio of a closed curve, and is, therefore, a firm result. Bound ii), equation (31), is realised for the $3_2 12_1$ tiling with geometry defined by equation (29). This is less firmly established and has the status of a conjecture. It may well be beaten by more complex periodic tilings, *e.g.*, composed of more than two different types of Plateau cells, but this remains an open question.

Another interesting problem that we have not addressed is whether there is an upper bound to the R of an unbounded, unstrained cluster. This would necessitate a more thorough investigation.

Finding lower (and upper) bounds for the surface energy of a 3d foam is of course much harder, and the answers are not known with certainty even in the monodisperse case. Here again progress has been guided by conjectures, starting with the seminal work of Kelvin [7]. At present, the strongest candidate minimiser is the Weaire-Phelan structure [8], which contains two types of cells of the same volume.

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