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Adaptation of the Z-cone model to the estimation of the energy of a bcc foam

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The Z-cone model, which provides an analytic approach to the estimation of the energy of a foam for simple crystal structures, has been adapted to the important case of the body-centred cubic (bcc) foam. This involves defining *two* types of cones, corresponding to the two sets of neighbouring bubbles that come into contact. We obtain results which compare favourably with accurate computations using the Surface Evolver. The precise dependence of energy on liquid fraction close to the loss of the $\langle 1\,0\,0\rangle$ contacts remains to be resolved.

Keywords: foams; soft matter; emulsions

1. Introduction

In the physics of foams [1–3], the structure envisaged by Kelvin [4, 5] has played a canonical role as a prototype, even though it is now known not to be the structure of lowest energy for a monodisperse dry foam [6, 7], as originally thought. The Kelvin structure consists of a body-centred cubic (bcc) arrangement of bubbles, and is stable with respect to small deformations for liquid volume fractions ϕ up to about $\phi \approx 0.11$ [8].

Various authors have already applied Surface Evolver simulations [9] to the dry Kelvin structure [8, 10–13]: in particular, Höhler *et al.* have used it when discussing foam structure for finite liquid fraction [15].

In this paper we show that the recently introduced analytical Z-cone model [16, 17] can be extended to a more general cone model, applicable to this case. Various approximations are involved in the new formulation, but no adjustable parameters. Part of our motivation was directed towards the details of the variation of energy close to the liquid fraction at which contact is lost with the six neighbours in the $\langle 1\,0\,0\rangle$ directions (*i.e.* the $\pm x$, $\pm y$ and $\pm z$ directions [18]). This may be expected to be related to the non-analytic forms found at the wet limit in various previous studies, including our previous Z-cone analysis [16, 17]. Such subtle questions are difficult to pursue with Surface Evolver simulations, due to the high level of precision required, and it was expected that the extended cone model would shed some light on the matter. Among other things, it should bear on the precise position and nature of the instability associated with the loss of $\langle 1\,0\,0\rangle$ contacts.

It is worth recalling what Kelvin himself was able to do at the outset [19]. He

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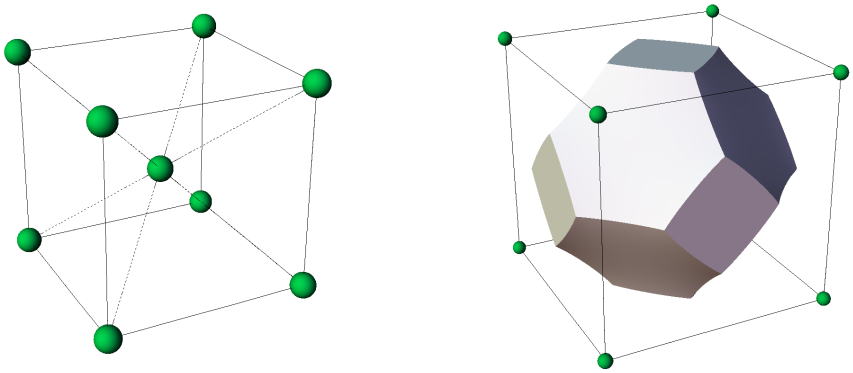


(a) (b)

Figure 1. (a) The bcc lattice. (b) A bubble in a dry bcc foam takes the form of a Kelvin cell. The hexagonal faces are slightly warped, and the square faces are planar with convex edges. (Produced using the Surface Evolver [9]).

was concerned only with the dry foam ($\phi = 0$), for which he produced a remarkably accurate description of the bubble shape: the *Kelvin cell* (Fig. 1). He recognised the implications of crystal symmetry: that the quadrilateral faces are flat, while the hexagonal ones are not, and applied Plateau's rules for the angles of intersection of faces, together with the requirement that the total curvature of the hexagonal faces is everywhere zero. His numerical calculation by hand of the approximate form of the hexagonal faces was a veritable *tour de force*.

But Kelvin did not proceed to estimate the surface area of his new structure, even though this bore directly on the motivation for the work. It appears that this was first evaluated one hundred years later, when Princen and Levinson [20] computed the surface area by using a discretisation into flat segments.

The result was expressed both in terms of the relative surface area S/S_0, where S_0 is the surface area of a sphere with the same volume as the bubble. The computed value of S/S_0 for the Kelvin cell is 1.0970, a decrease from the value of 1.0990 for the planar-faced truncated octahedron, which to Princen and Levinson appeared "surprisingly small" [20]. A simple estimate, based on adjusting angles of the truncated octahedron to conform with Plateau's rules, gives $S/S_0 = 1.0968$: details are given in Appendix A.

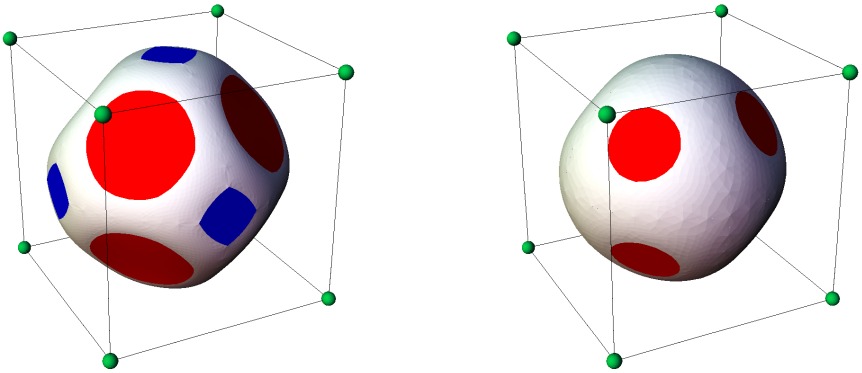
Here we will express energies in terms of the *dimensionless excess energy* ε, defined as

$$\varepsilon(\phi) = \frac{S(\phi)}{S_0} - 1. \tag{1}$$

At the dry limit, we have $\varepsilon_0 = 0.0970$ (obtained from Surface Evolver simulations, in agreement with [20]. At the wet limit, $\varepsilon(\phi_c) = 0$.

One motivation for revisiting the bcc foam is that it is a well-defined structure that can be used for the study of a general feature of foams: namely, the gain or loss of a face at some critical liquid fraction. At the outset, it is not obvious that the Z-cone model can be readily adapted to this case, in which all neighbours are not equivalent; it turns out that this can indeed be achieved.

As mentioned earlier, for $\phi \gtrsim 0.11$ the Kelvin foam becomes unstable, and close-



(a) (b)

Figure 2. Two examples of equilibrium bubble shape in a wet bcc foam, with centres of neighbouring bubbles marked. (a) For liquid fraction $\phi < \phi^*$ ($\phi = 0.05$ here) there are two sets of contacts, corresponding to the (dry) hexagonal $\langle 1\,1\,1 \rangle$ and square $\langle 1\,0\,0 \rangle$ faces. (b) When ϕ exceeds ϕ^* ($\phi = 0.15$ here)the square contacts are lost. (Produced using the Surface Evolver [9]).

packed structures have a lower energy. Nonetheless, here we discuss the energy of a bcc-ordered foam over the full range of liquid fraction, in ways that do not allow instability.

As we increase ϕ from 0 to its maximum value of ϕ_c, at which bubbles are spheres ($\phi_c = 1 - \sqrt{3}\pi/8 \approx 0.32$), the square contacts corresponding to $\langle 1\,0\,0 \rangle$ neighbours shrink, and disappear at a liquid fraction ϕ^*: see Fig. 2. The association of this loss with instability dates back to some incidental remarks of [21], inspired by the instability of bcc metals.

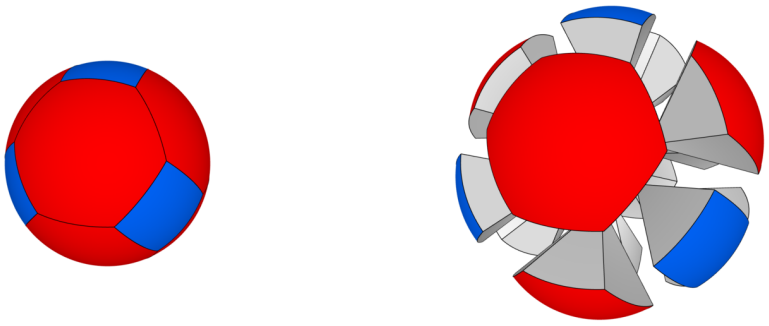
2. The wet foam

Here we present an approximation, based on a generalisation of the recently introduced Z-cone model, and compare our results to accurate simulations using the Surface Evolver. The latter were carried out using symmetry considerations, similar to those in [22]: a fuller description of the simulations is presented in Appendix B.

2.1. *The cone model applied to the Kelvin foam*

The Z-cone model is an analytically tractable model that allows for the estimation of the energy of a foam consisting of identical bubbles with Z nearest neighbours [16, 17]. In the original Z-cone model each bubble is segmented into Z equivalent pieces which are then approximated as circular cones. Upon deformation (corresponding to the compressed packing of bubbles in a foam) the initially spherical caps of the cones become increasingly flattened. Their surface area is minimised subject to a specified cone volume. The result is an analytic parametric expression for the excess energy ε of a bubble in terms of liquid fraction ϕ which contains no free parameters and depends only on the number of contacts. For full mathematical details see our previous papers [16, 17].

In order to deal with the Kelvin cell we have extended the Z-cone model to comprise two different types of cones: see Fig. 3. Eight identical cones of specified



(a) (b)

Figure 3. (a) A bubble in a Kelvin foam (represented here as a sphere) has eight $\langle 1\,1\,1\rangle$ neighbours (hexagonal faces) and six $\langle 1\,0\,0\rangle$ neighbours (square faces). (b) We associate with each neighbour a cone, as shown here for a spherical bubble. In the cone model, each of these is approximated as a *circular* cone. The total surface area of the caps of the cones is minimised subject to appropriate constraints, including a fixed total volume when bubbles are brought into contact and become distorted.

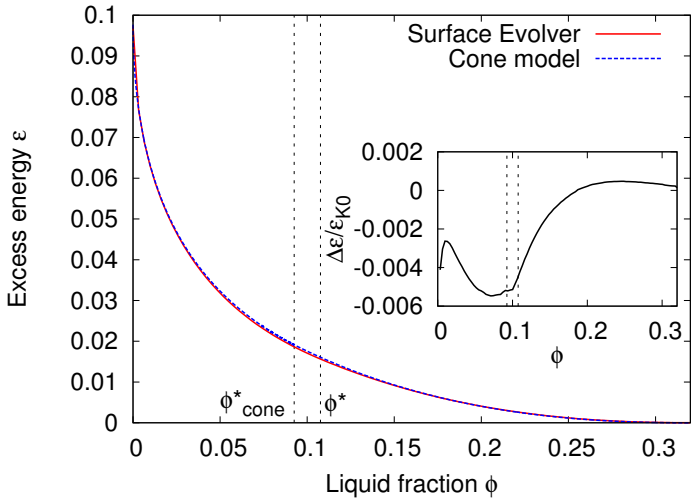


Figure 4. Variation of dimensionless excess energy ε (as defined in Eq. (1)) with liquid fraction ϕ for the Kelvin structure, obtained from Surface Evolver calculations (solid line), and its approximation using the generalised cone model (dashed line). The values in the dry limit ($\phi = 0$) are $\varepsilon_0 = 0.0970$ from the Surface Evolver and $\varepsilon_{0,C} = 0.0980$ from the cone model. Increasing ϕ leads to the loss of the six square faces. This takes place at $\phi^* = 0.108$ for Surface Evolver simulations, and at $\phi^*_{\rm cone} = 0.092$ in the cone model: see dashed vertical lines on plot. The inset shows the normalised difference $(\varepsilon(\phi) - \varepsilon_{\rm cone}(\phi))/\varepsilon_0$.

opening angle correspond to the eight hexagonal faces and six identical cones, with a different opening angle, correspond to the six square faces. Appropriate constraints relate the solutions for the two types of cone, to ensure that they match appropriately at boundaries. For a detailed description of the extended cone model see Appendix C.

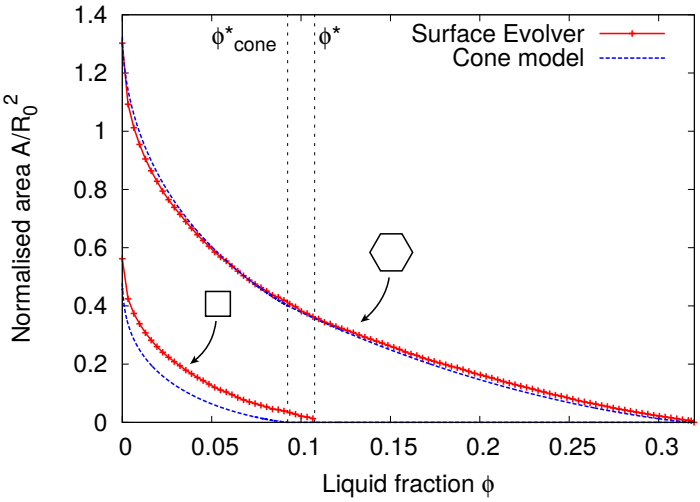


Figure 5. Variation of the areas of hexagonal and square contacts with liquid fraction, obtained from the Surface Evolver and the cone model. The areas are normalised by R_0^2, where the bubble volume is $4/3\pi {R_0}^3$.

2.2. *Results*

Fig. 4 shows the variation of the dimensionless excess energy $\varepsilon(\phi)$ with liquid fraction, obtained from both Surface Evolver and the cone model — a surprisingly good estimation of the excess energy over the entire range of ϕ, with the difference not exceeding one percent of $\varepsilon_0 = \varepsilon(0)$. It is worth noting that we see better agreement here than in the fcc case of the original Z-cone model [16]. This is due to the fact that we approximate the contacts as circles: the square and hexagonal contacts in a bcc foam are more rotationally symmetric than the rhombuses of an fcc foam. Similarly, for $Z = 12$ in the original Z-cone model we saw much better agreement with simulation for a pentagonal dodecahedron than the rhombic dodecahedron [16].

The value of ϕ at which the square $\langle 1\,0\,0\rangle$ contacts vanish is given as $\phi^* = 0.108$ by the Surface Evolver, and $\phi^*_{\text{cone}} = 0.092$ by the cone model. (It is worth noting that Weaire *et al.* [21] arrived at a remarkably accurate early estimate of $\phi^* \approx 0.11$ by a crude argument based on ratios of Plateau border widths.) The critical liquid fraction for the wet limit is $\phi_c = 0.320$ for the Kelvin foam; the cone model arrives at a good approximation $\phi_{c,\text{cone}} = 0.319$.

Fig. 5 shows the variation in area of both square and hexagonal faces (recall that it is the loss of the former that corresponds to ϕ^*). When ϕ is very slightly less than ϕ^* we encounter problems in accurately modelling the surface using the Surface Evolver, due to difficulties in allowing the area of facets to go to zero.

We now give closer attention to the two critical points, ϕ^*_{cone} and $\phi_{c,\text{cone}}$. In doing so, results are clearer when viewed in terms of derivatives. We show in Fig. 6 the variation of the derivative $\mathrm{d}\varepsilon/\mathrm{d}\phi$ with liquid fraction as obtained from the cone model. The asymptotic behaviour of $\mathrm{d}\varepsilon/\mathrm{d}\phi$ near the wet limit $\phi = \phi_{c,\text{cone}}$, as obtained from differentiating the result from the original Z-cone model [16], and keeping the highest-order term:

$$\frac{\mathrm{d}\varepsilon}{\mathrm{d}\phi} \sim a\frac{\phi_{c,\text{cone}} - \phi}{\log(\phi_{c,\text{cone}} - \phi)}, \tag{2}$$

where a is a constant [16]. This describes the present data well, as one might expect: see Fig. 6(b).

For $\mathrm{d}\varepsilon/\mathrm{d}\phi$ at $\phi = \phi^*_{\mathrm{cone}}$ we did not succeed in finding an analytical expression from our new cone model, on account of the numerical procedures involved (see Appendix C). The following empirical expression is a reasonably good description of the data near $\phi = \phi^*_{\mathrm{cone}}$ (see Fig. 6(c)):

$$\frac{\mathrm{d}\varepsilon}{\mathrm{d}\phi} \sim b_1 + \frac{b_2}{(\log(\phi^*_{\mathrm{cone}} - \phi))^2}, \tag{3}$$

with two parameters $b_1 < 0$ and $b_2 > 0$.

There is a discontinuity of the slope of $\mathrm{d}\varepsilon/\mathrm{d}\phi$ at $\phi = \phi^*_{\mathrm{cone}}$, which is clearly visible in Fig. 6(c).

Of note is the presence of logarithmic terms in both expressions, a feature known from various studies of bubble-bubble interactions [23, 24]. The discrepancy between the two forms (2) and (3) suggests that results from bubble-bubble interactions do not directly apply to all contact losses away from the wet limit, but we have no explanation for this difference.

It has been argued that the limit of mechanical stability of the Kelvin structure is directly attributable to the loss of the square faces [25]. A bcc crystal of interacting points is well known to require second-nearest-neighbour interactions to stabilise it when simple pairwise potentials are applied. [26, 27].

This appeared indeed supported by Phelan *et al.* [8], who found a negative elastic constant at values of $\phi > 0.11$, *i.e.* very close to the value of $\phi = 0.11 \pm 0.005$ that these authors identified for the face loss. New preliminary Surface Evolver calculations by us are in accord with this — however, such an analysis is complicated by the anomalous variation of energy at this critical point; this requires to be examined more closely.

3. Conclusions

We have successfully extended the Z-cone model to the Kelvin foam, where not all contacts are equivalent. The energy computed from this model agrees very well with Surface Evolver simulations over the entire range of liquid fractions.

We have examined two distinct contact losses: the loss of the hexagonal $\langle 1\,1\,1 \rangle$ faces at the wet limit, ϕ_c, and of the square $\langle 1\,0\,0 \rangle$ faces away from the wet limit at ϕ^*, resulting in two distinctly different variations of energy with liquid fraction. We previously derived an expansion for $\mathrm{d}\varepsilon/\mathrm{d}\phi$ near $\phi = \phi_c$ which revealed a logarithmic dependence; we now have an empirical expression for the same derivative near $\phi = \phi^*$ which, too, features a logarithmic term.

Using the cone model, we have also computed the variation of the areas of the hexagonal and square faces close to ϕ_c and ϕ^* respectively: numerical noise hinders these calculations using the Surface Evolver.

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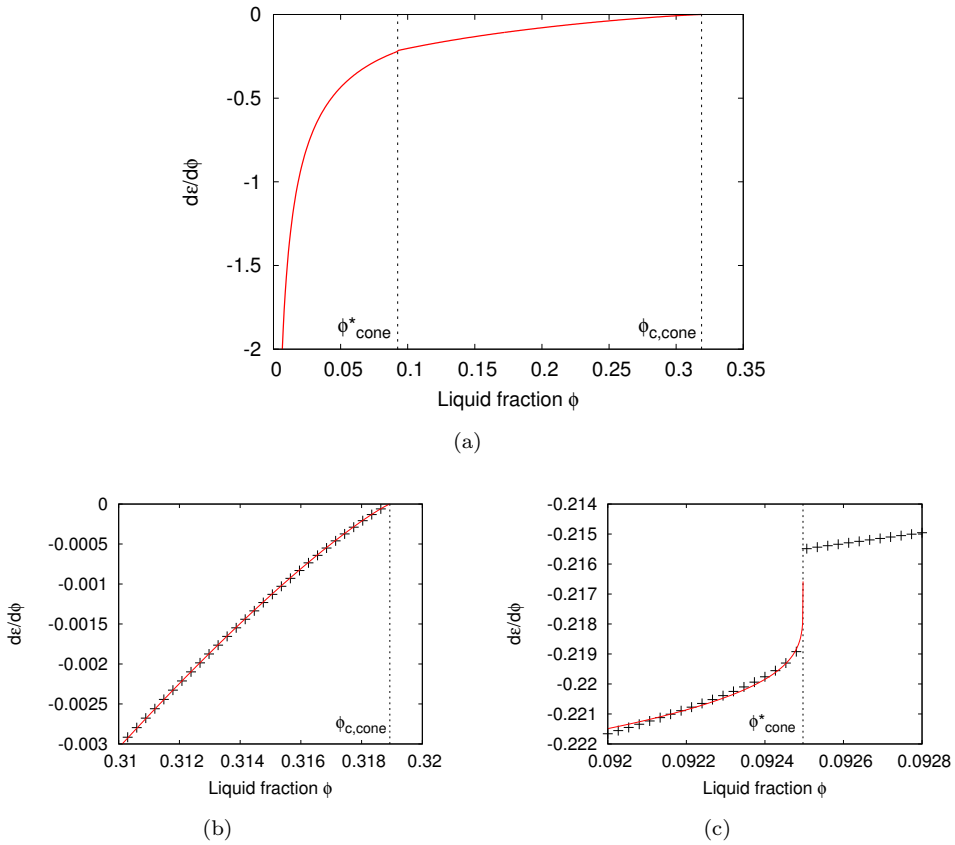

Figure 6. (a) The derivative of excess energy with respect to liquid fraction, $\mathrm{d}\varepsilon/\mathrm{d}\phi$, computed numerically from the cone model. There is a discontinuity of slope at $\phi = \phi^*_{\mathrm{cone}} \approx 0.092$, *i.e.* the point of next-nearest neighbour contact formation/loss (square faces). (b) Near $\phi = \phi_{c,\mathrm{cone}}$ (the wet limit), the variation of the derivative $\mathrm{d}\varepsilon/\mathrm{d}\phi$ (points) is well approximated by the form in Eq. (2) (continuous line), obtained from the simple Z-cone model. (c) Near $\phi = \phi^*_{\mathrm{cone}}$ (loss of square faces), the variation is quite different, and is reasonably well approximated by the proposed empirical form of Eq. (3) (continuous line).

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Appendix A. Estimating the energy of the dry Kelvin foam

In the course of this work, we have noticed that the energy of the dry Kelvin foam can be well estimated in a very elementary way, which may have applications to other cases.

The natural first approximation to the Kelvin cell is the Voronoi cell of the bcc lattice: the truncated octahedron, with fourteen flat faces which are planes equidistant from first and second neighbour bubbles. The angles between these planes do not conform to Plateau’s equilibrium rules, that is, they are not 120°. The equilibrium structure therefore has slightly lower energy. Our objective is to estimate the reduction in energy when the Voronoi structure is relaxed.

We first note that the Voronoi structure can be held in equilibrium by applying

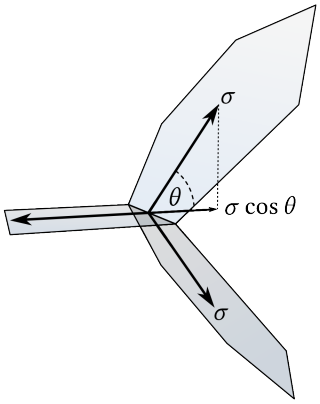


Figure A1. If a surface tension is associated with the faces of the truncated octahedron, it is not in equilibrium. This sketch shows the forces due to surface tension σ acting at an edge between a quadrilateral face and two hexagons. Note that in the unrelaxed truncated octahedron 2θ is simply the dihedral angle of a regular octahedron, *i.e.* $2\theta = \arccos(-1/3) \approx 109.47°$.

additional external forces at the edges of the quadrilateral faces, to compensate for the mismatch of surface tensions (see Figure A1).

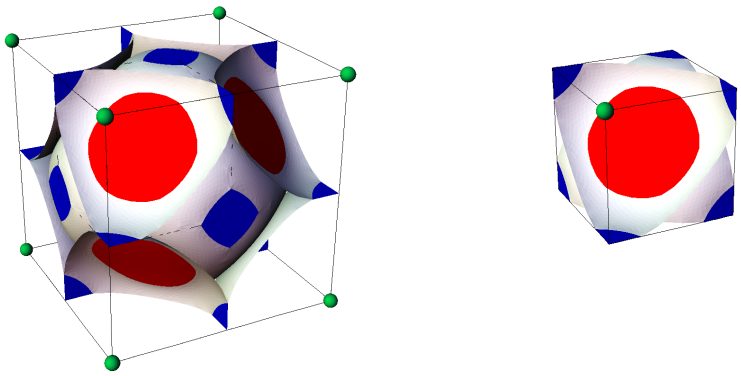
We then proceed to estimate the (negative) work done by these fictitious forces as they are continuously reduced to zero. For each increment of such a change, the work is simply force times displacement. To incorporate the latter, we approximate the curved edges of the square faces as parabolic, and the force as conforming to Hooke's law. An elementary calculation then gives $S/S_0 = 1.0968$.

Kusner and Sullivan sketch the computation of a *lower bound* for the energy of a Kelvin cell in [11] using similar arguments as above, resulting in $S/S_0 \geqslant 1.0954$. The full details appear to be unpublished.

Appendix B. The bcc structure and the Surface Evolver

Fig. B1(a) shows the conventional bcc cell for a foam. The Kelvin foam consists of repeated translated copies of this cell. However, for simplicity we can exploit some of the symmetries of the conventional cell: namely, reflectional symmetry in the x, y and z directions (Brakke and Sullivan [22] exploit even more symmetries to yield a minimal representation of the fully dry Kelvin foam). Hence we arrive at a reduced cell (Fig. B1(b)), which has one eighth of the volume of the conventional cell, and is composed of a cube containing one eighth of a bubble at each of two opposite corners. This increases the speed of computation considerably.

We begin with a very roughly triangulated approximation of the configuration in Fig. B1(b), with appropriate film edges constrained to lie within the faces of the cube, *i.e.* planes of reflection. Minimisation of energy results in films meeting the faces of the cube at 90°, which ensures that the resulting foam structure is smooth. We note that in order to faithfully represent the full foam, films which lie *within* these planes (in this case, the blue $\langle 1\,0\,0 \rangle$ faces) are given half of their 'real' surface tension. Hence we give the red $\langle 1\,1\,1 \rangle$ contact face a tension of 2 and all other facets a tension of 1. Iterated mesh refinements and gradient-descent minimisations yield the configuration shown in Fig. B1(b): the same surface is visualised as a single



(a) (b)

Figure B1. (a) The equilibrium structure for a Kelvin foam, including all surfaces within the conventional cell (*c.f.* Fig. 2 which shows the surfaces of a single bubble). The $\langle 1\,1\,1\rangle$ contact faces are shown in red, and the $\langle 1\,0\,0\rangle$ faces in blue. (b) We exploit reflectional symmetries to obtain a representative cell one eighth the size of (a). The full foam can be built from reflected and translated copies of the representative cell.

bubble in Fig. 2(a).

Appendix C. The cone model applied to bcc

Modelling the Kelvin cell with the cone model may be regarded as a first step in extending this approach to more general ordered foam structures in which not all of the contacts are equivalent. The presence of two different types of contacts in the bcc structure adds a geometric complexity to the cone model which means that several simplifying statements in the definition of the original model no longer hold. The goal of this section is to give some details of how these additional complications are incorporated.

While the total volume of the bubble, V_0, is constant, the volume of each of the cones is no longer required to be constant. The constraint on the individual cone volumes is now given by

$$8V_h + 6V_s = V_0,$$

where V_s and V_h denote the volumes of the cones associated with square and hexagonal bubble contact areas, respectively.

The second additional complication is the determination of the opening angles θ_h and θ_s of each type of cone (see Fig. C1). We choose to retain the values of the solid angles subtended by each type of face in the "dry" Kelvin structure. This ensures that the sum of the solid angles subtended by the eight hexagonal and six square faces is equal to the 4π steradian solid angle of our bubble.

Two cones which meet each other are required to have a common slant height r_s (see Fig. C1) so that their curved caps match. In the original Z-cone model (with identical cones) we required each of the curved caps to meet their respective cones at right angles. In the case of the bcc bubble, this is more subtle. As can be seen in Fig. 3, a square cone joins only with four hexagonal cones (with corresponding angle γ_s), whereas a hexagonal cone joins with three square cones and three hexagonal

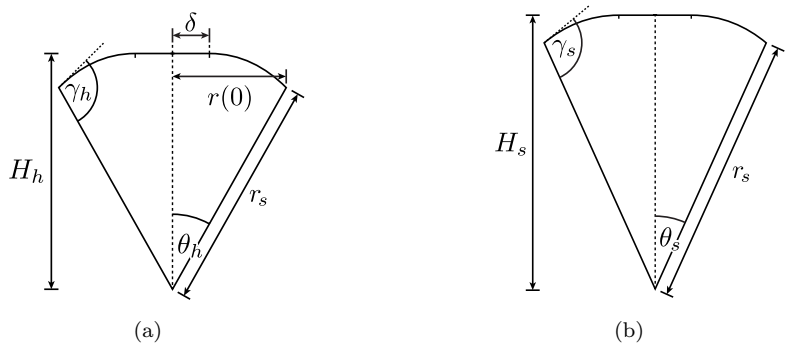


Figure C1. In the extended cone model we deal with *two* types of cones: (a) for the hexagonal ⟨1 1 1⟩ contacts and (b) for the square ⟨1 0 0⟩. They share a common slant height r_s. The ratio $\delta/r(0) = \rho_\delta$ (where δ is the radius of the contact, and $r(0)$ the maximum width of the cone, as shown) features in the derivation of the cone model expressions.

(with corresponding angles γ_{hs} and γ_{hh} respectively). Smoothness requires $\gamma_{hh} = \pi/2$, and that

$$\gamma_s + \gamma_{hs} = \pi \tag{C1}$$

The cone model requires a single angle, γ_h, which we define as an average $\gamma_h = (\gamma_{hh} + \gamma_{hs})/2$. Hence we can rewrite (C1) as

$$2\gamma_h + \gamma_s = \frac{3\pi}{2}.$$

Similarly to the generalised volume condition above, the angles γ_h and γ_s are no longer fixed, as in the Z-cone model. We can make use of the following constraints to uniquely determine V_h, θ_h and γ_h for any given liquid fraction ϕ.

The first constraint we impose is that the ratio ν of cone heights is constant with respect to liquid fraction. Adopting the solid angles from the dry Kelvin structure, and requiring that cone slant heights match, fixes this ratio:

$$\nu = \frac{H_h}{H_s} = \frac{\cos\theta_h}{\cos\theta_s} = 0.8644. \tag{C2}$$

Note that for a real Kelvin foam the corresponding ratio is $\sqrt{3}/2 \approx 0.866$.

The final constraint on our problem is that the internal pressure p in each of the neighbouring cones should be equal. This is simply the statement that pressure does not depend on the position in the bubble. The internal pressure of a bubble is responsible for the curvature of its surface and, by considering the work done to increase the volume of each cone by a small amount ΔV_i, while keeping the size of the contact constant (*i.e.* blowing it up slightly), we arrive at the following expression for the internal pressure of a cone,

$$p_i = \frac{\Delta E_i}{\Delta V_i^*} - 2\pi r_{s_i} \cos\gamma_i \cos\theta_i \frac{\Delta r_{s_i}}{\Delta V_i^*}, \tag{C3}$$

where Δr_{s_i} is the slant height change of a cone, ΔE_i is the surface energy change

and ΔV_i^* the change in the volume associated with the curved surface of the bubble. The first term represents the work necessary to increase the free surface of the cone by an amount ΔV_i^*, while the second is the work done by the surface tension σ in changing the surface energy of the cone to account for the increase in slant height. It can be thought of in terms of reduction of curvature:

$$W_{\sigma_i} = 2\pi r_{s_i} \Delta r_{s_i} \cos\theta_i \cos\gamma_i.$$

The above constraints are sufficient to determine all of the variables in our problem and to write the excess surface energy ε for the Kelvin cone model as

$$\varepsilon(\rho_{\delta_h}, \rho_{\delta_s}, \theta_h, \theta_s, \Gamma_h, \Gamma_s, q_h, q_s) = \frac{8A_h(\rho_{\delta_h}, \theta_h, \Gamma_h, q_h) + 6A_s(\rho_{\delta_s}, \theta_s, \Gamma_s, q_s)}{4\pi} - 1.$$

Here, the area of each face is given by

$$A_i(\rho_{\delta_i}, \theta_i, \Gamma_i, q_i) = \pi R_0^2 \left(\frac{4q_i}{3J(\rho_{\delta_i}, \Gamma_i) + \cot\theta_i}\right)^{\frac{2}{3}} \left[\rho_{\delta_i}^2 + 2(1-\rho_{\delta_i}^2)K(\rho_{\delta_i}, \Gamma_i)\right],$$

and the angles Γ_h and Γ_s are related to γ_h and γ_s via

$$\Gamma_i = \gamma_i + \theta_i - \frac{\pi}{2}.$$

The quantities q_s and q_h are fractions of the total volume V_0 taken up by any one of the square or hexagonal cones:

$$q_s = \frac{V_s}{V_0}; \quad q_h = \frac{V_h}{V_0},$$

and ρ_δ is given by the ratio of lengths $\delta/r(0)$ as shown in Fig. C1(a).

The liquid fraction can be similarly expressed in these terms as

$$\phi(\rho_{\delta_h}, \theta_h, \theta_s, \Gamma_h, q_h) = 1 - \frac{3J(\rho_{\delta_h}, \Gamma_h) + \cot\theta_h}{2q_h\left[I(\rho_{\delta_h}, \Gamma_h) + \cot\theta_h\right]^3\left(4\tan^2\theta_h\left(\frac{3}{v^3}\right) + \tan^2\theta_s\right)}$$

The elliptic integrals are given explicitly by:

$$I(\rho_\delta, \Gamma_i) = \int_{\rho_{\delta_i}}^1 \sin\Gamma_i\left(x^2 - \rho_{\delta_i}^2\right) f(x, \rho_{\delta_i}, \Gamma_i)\,\mathrm{d}x,$$
$$J(\rho_\delta, \Gamma_i) = \int_{\rho_{\delta_i}}^1 \sin\Gamma_i x^2\left(x^2 - \rho_{\delta_i}^2\right) f(x, \rho_{\delta_i}, \Gamma_i)\,\mathrm{d}x, \text{ and}$$
$$K(\rho_\delta, \Gamma_i) = \int_{\rho_{\delta_i}}^1 x^2 f(x, \rho_{\delta_i}, \Gamma_i)\,\mathrm{d}x,$$

with

$$f(x,\rho_{\delta_i},\Gamma_i)=\left[x^2(1-\rho_{\delta_i}^2)^2-\sin\Gamma_i^2\left(x^2-\rho_{\delta_i}^2\right)^2\right]^{-\frac{1}{2}}.$$

For a more detailed derivation of the original Z-cone model, see [17].

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