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Bubble-bubble interactions in a 2d foam, close to the wet limit[☆]D. Weaire^a, R. Höhler^{b, c}, S. Hutzler^{a, *}^a School of Physics, Trinity College Dublin, The University of Dublin, Ireland^b Université Paris-Est Marne-la-Vallée, 5 Bd Descartes, Champs-sur-Marne, F-77454 Marne-la-Vallée cedex 2, France^c Sorbonne Universités, UPMC Université Paris 06, CNRS-UMR 7588, Institut des NanoSciences de Paris, 4 place Jussieu, Paris 75005, France

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ABSTRACT

Following the general approach of Morse and Witten for the deformation of a bubble in contact with neighbouring bubbles, we develop a model for contacting bubbles in two dimensions which can be solved analytically. The force-displacement relations are derived by elementary methods; unlike the case of 3d, no logarithmic factors arise in two dimensions. We also discuss the case of a uniform compression of a symmetric foam structure; the (osmotic) compressibility depends on the number of contacts, as was shown in earlier work by Lacasse et al. Our model, which is based on first principles, without any free parameters, may be extended to simulate 2d foams.

1. Introduction

In the theory of liquid foams [1,2] the idealized two-dimensional model has played an important role. It can represent some real systems, such as a layer of bubbles trapped between plates [3]. It also serves to expose and illustrate many properties which are also characteristic of three-dimensional foams, for which they may be much more difficult to visualize and analyze. Theoretical results often take simpler forms in two dimensions.

In the present case we seek to understand the interactions between 2d bubbles close to the wet (or jamming) limit, where they become circular and the structure loses its ability to resist a static stress (rigidity loss transition [4]). This problem has not been studied intensely for as long as that of the dry limit, where the 2d bubbles take the form of polygons with curved sides; see Fig. 1.

Much of what has been advanced in describing wet foams has been based on a simple *ad hoc model* in which 2d bubbles are represented by circles (or spheres in three dimensions) whose overlap is resisted by a force which varies linearly with distance between the centres of the circles [8–11]. This “Bubble Model”, used also extensively in studies of the jamming transition for soft particles [12], has been a valuable guide to general trends of foam properties, such as

energy or shear modulus. It is convenient for computation but it is clearly unreliable in detail.

What then is the true nature of the interaction between contacting bubbles? Morse and Witten [13] undertook to answer the question in the case of three dimensions, in a now classic paper. They raised it in the context of emulsions but for present purposes this makes no difference. The equilibrium properties of both bubbles and emulsion droplets are generally well described in terms of incompressible fluids and a constant surface tension.

The analysis of Morse and Witten was highly original and penetrating, but difficult to follow in detail, so that it is still poorly appreciated even today. Only a few papers have addressed it, or developed it further, e.g. [14–17]. The present paper arises from an attempt to thoroughly rework the Morse-Witten analysis, in the course of which it became clear that the simpler two-dimensional case, hitherto neglected, was worthy of attention. Empirical work on bubble interactions, based on experimental data, exists only for quasi-two-dimensional emulsion droplets [18].

The main qualitative result of Morse and Witten was surprising: that the energy E associated with the deformation of two contacting bubbles or a bubble contacting a wall, written as a function of the force F between them, involved the logarithm of F . To lowest order in

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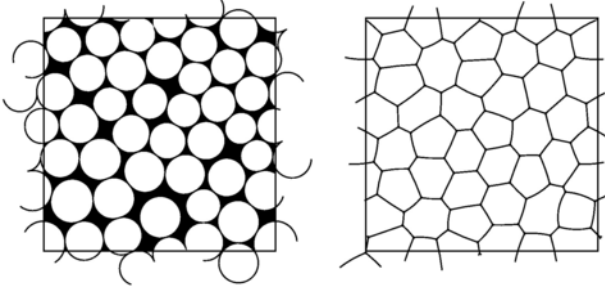


Fig. 1. Examples of two-dimensional foam structures in the wet (near-circular bubbles) and dry regimes (near polygonal bubbles), obtained using the software PLAT [5–7].

F , their result is

$$E \sim F^2 \ln(F^{-1}). \quad (1)$$

A corresponding result, confirmed by simulations [19] and related theories [20,21], as well as experiments [16], may also be written in terms of force and displacement, also involving a logarithm [20]. It deserves to stand among other canonical forms of force laws in nature, such as that of Hertz for elastic particles [22], or Hooke's law.

What is the form of the corresponding interaction in two dimensions? Can it be adapted and extended in the manner of Höhler and Cohen-Addad [17] to create a formalism of pairwise interactions, to model energy, elastic moduli, etc.? We address these questions here. Our initial analysis of a single bubble subject to an applied force will be much more direct and elementary than that used by Morse and Witten [13] for their 3d problem. A detailed review of the latter will be presented in a further paper [23].

2. Single 2d bubble subject to a single contact force

The unperturbed bubble is taken to have radius R_0 . Its area will be conserved. This is a familiar condition, corresponding in practice to the incompressibility of the contained gas, under the weak forces encountered in many situations where the Laplace pressure is many orders of magnitude smaller than the bulk modulus of the gas, and where there is no significant pressure gradient [2,24].

We wish to find the profile of the bubble under the action of a single point force of magnitude F , but in order for equilibrium to be possible, it must be opposed by another. Following the lead of Morse and Witten [13], we introduce an equal and opposite uniform body force via an internal pressure gradient $F/(\pi R_0^2)$ per area. In an experiment, with a layer of bubbles or droplets confined between two parallel plates, this body force could be implemented by simply by tilting the plates, as in the experiments of Desmond et al. [18].

The profile of the deformed bubble, Fig. 2, will be expressed in (dimensionless) circular polar coordinates r and θ ,

$$r(\theta) = 1 + \delta r(\theta), \quad (2)$$

where $\delta r(\theta)$ is the (dimensionless) bubble displacement, see Appendix A.

For small radial displacement $\delta r(\theta)$, it is governed by the linear differential equation

$$-\left(\frac{d^2}{d\theta^2} + 1\right) \delta r(\theta) = a + \frac{f}{\pi} \cos \theta \quad (3)$$

where f is a dimensionless force, given by $f = F/\gamma$, and γ is the sur-

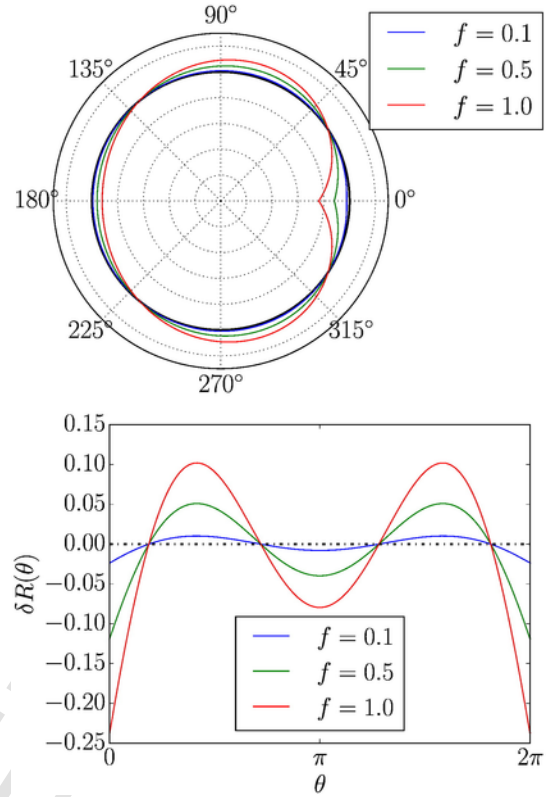


Fig. 2. Shape of a two-dimensional bubble in contact with a (dimensionless) point force f acting on its surface at angle $\theta = 0$. (a) The thick black line represents the undeformed circular bubble of the same area (radius $r(\theta) = \text{const.} = 1$). The coloured lines show the displacement $\delta r(\theta)$ in response to point forces $f = 0.1, 0.5, 1.0$. (b) Radial displacement $\delta r(\theta)$ as a function of θ . (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

face tension for a gas-liquid interface in 2d; see Appendix A. The right-hand side is the pressure variation associated with the compensating force field mentioned above. The constant a will be specified below.

Eq. (3) is the 2d equivalent of Eq. (3) of Morse and Witten [13], and may be obtained by linearising the full expression for curvature in the circular polar coordinates, and writing the condition for local equilibrium of the curved edge of the bubble, which involves surface tension (hence curvature) and pressure. This is the Laplace-Young law; see Appendix A.

The general solution of Eq. (3) is elementary:

$$\delta r(\theta) = \left(c_1 - \frac{f}{2\pi}\right) \cos \theta + \left(c_2 - \frac{f}{2\pi}\theta\right) \sin \theta - a. \quad (4)$$

It includes two arbitrary constants c_1 and c_2 in addition to a .

The constant c_1 is determined by demanding that the centre of mass is not shifted, as in the 3d solution derived by Morse and Witten [13]. The condition

$$\int_0^\pi \delta r(\theta) \cos \theta d\theta = 0 \quad (5)$$

then results in $c_1 = f/(4\pi)$. Later we will represent the equilibrium structure of a 2d foam in terms of a network of forces: their lines of action will be taken to meet in a point for each bubble. This is justifiable, to within a negligible order; the point is the centre of mass [23].

Using the obvious symmetry of the problem we may apply the condition $\frac{d\delta r}{d\theta} = 0$ at $\theta = \pi$ in order to determine $c_2 = f/2$.

Finally the arbitrary constant a , related to the internal pressure, is determined by the condition of incompressibility, which is

$$\int_0^\pi \delta r(\theta) d\theta = 0. \quad (6)$$

This gives $a = f/(2\pi)$, hence the full solution that we require is,

$$\delta r(\theta) = \frac{f}{2\pi} \left[(\pi - \theta) \sin \theta - \frac{\cos \theta}{2} - 1 \right], \quad (7)$$

plotted in Fig. 2 for three different values of force f . Note that unlike the situation in 3d [13], here the deformation is not singular where the point force is applied.

A useful check is to consider force and surface tension at the point where the force is applied (at $\theta = 0$ in Fig. 2). Computation of the slope of the bubble profile at this point shows that these are indeed in equilibrium.

In Section 4 we will demonstrate the validity of the model for the case of a bubble compressed between two plates, where an analytical solution exists.

3. Introduction of a finite contact

In relation to foams the case of a point contact is not directly relevant, but it can be adapted, following the lead of Morse and Witten. This introduces a flat ‘cap’ as shown in Fig. 3, representing the line of contact between two bubbles of equal pressure, with no change of the force f , now distributed uniformly on it.

Since the internal pressure (dimensionless Laplace pressure) is equal to unity, in lowest order, the corresponding estimate of the

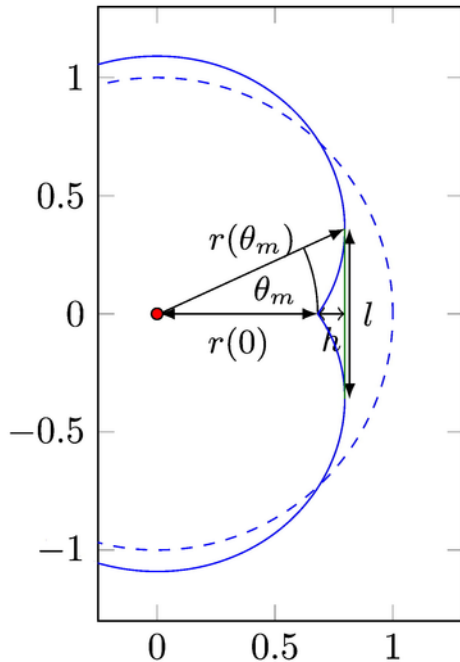


Fig. 3. Sketch showing the flat ‘cap’ forming at the line of contact between two bubbles. (For purpose of illustration we are showing the case of a large deformation.) Expressions for cap length l and height h as a function of force f are given in Appendix B. In the 3d case, part of the corresponding profile is unphysical, essentially because it violates the assumption that δr is small (instead it diverges). In 2d this is not the case, for small f .

length of the cap is f . Appendix B includes the details of the derivation of its position, by an elementary method. This leads to two important conclusions, as follows. Firstly, the displacement at $\theta = 0$ is found to be linear in f to first order. Secondly, the change in area of the enclosed curve, consequent upon introducing the cap, is of higher order, $\mathcal{O}(f^3)$, than f , and need not be compensated.

It follows that, although we may have in mind the capped profile, the original one can be used in what is to come, within a lowest order approximation, in expressing displacement (with the centre of mass fixed) as a function of angle.

4. Force-displacement relations for several contacts

We now consider how this theory may be applied to an assembly of contacting bubbles, to arrive at a formalism similar to that used by Höhler and Cohen-Addad in 3d [17], which consists of explicit relations between force and displacement where all the contacts of a bubble are coupled to each other, due to the conservation of the bubble volume.

The radial displacement $\delta r(\theta)$, Eq. (7), is a linear function in f , periodic in 2π . We will write it as $\delta r(\theta) = \frac{f}{2\pi} g(\theta)$, with

$$g(\theta) = (\pi - \theta) \sin \theta - \frac{\cos \theta}{2} - 1. \quad (8)$$

Proceeding in parallel with the formulation of Morse and Witten (see also [23]), the displacement at contact i is given by the following sum over all contacts j that a bubble has,

$$\delta r_i = \frac{1}{2\pi} \sum_j f_j g(\theta_{ij}). \quad (9)$$

Here $g(\theta_{ij})$ takes the role of a Green's function (corresponding to Eq. (8) of Morse and Witten [13]) and θ_{ij} is the angle between the vectors \vec{r}_i and \vec{r}_j which point from the centre of the undeformed bubble to contacts i and j , respectively.

As an illustration of the theory and the above relationship we will consider the case of a bubble which is compressed between two parallel plates, with contacts at angles 0 and π , as in Fig. 4. Its shape is described by $r(\theta) = 1 + \delta r(\theta) + \delta r(\theta + \pi)$. For a deformation less than about 10 percent this turns out to be a very good approximation of the exact result, which consists of two semi-circles meeting both top and bottom plates tangentially, separated by a rectangle. Fig. 4 (b) shows the deviations from the exact shape for a deformation of 20%. Here the indentations of the bubble at the two contacts are very pronounced, as is the lateral bubble extension required for area conservation.

In order to obtain the force-displacement relationship it is sufficient, due to symmetry, to compute the displacement of the contact at angle 0 only, resulting in $\theta_{11} = 0$ and $\theta_{12} = \pi$. From Eq. (8) we then obtain $g(\theta_{11}) = -3/2$ and $g(\theta_{12}) = -1/2$, resulting in $\delta r_1 = -f_1/\pi$ by use of Eq. (9). Using symmetry, we obtain $\delta r_1 = \delta r_2 = -f/\pi$. Re-instating physical dimensions gives the force per contact as $F = -2\pi\gamma \frac{\delta R}{R_0}$.

It is straightforward to compute the total energy of this bubble without any approximations, since its shape consists of two semi-circles, separated by a rectangle (whose shorter sides make up the contact). From this the force per contact is evaluated as $F = -2\pi\gamma \frac{\delta R}{R_0} \left(1 + \frac{\delta R}{2R_0}\right) \left(1 + \frac{\delta R}{R_0}\right)^{-2}$, which agrees with the expression above to lowest order.

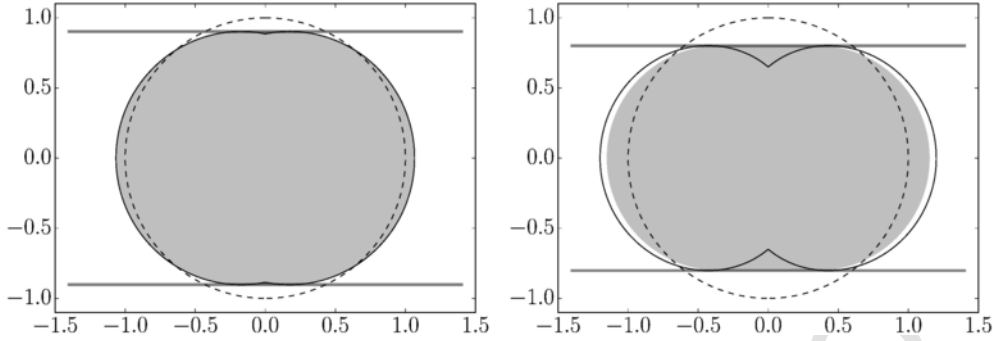


Fig. 4. Shape of a two-dimensional bubble in contact with two identical point forces f acting on its surface at angle $\theta = 0$ and $\theta = \pi$ (solid line). The dashed line represents the undeformed circular bubble, the grey area marks the exact solution and the solid line the solution $r(\theta) = 1 + \delta r(\theta) + \delta r(\theta + \pi)$ (see Eq. (7)). (a) In the case of the 10% deformation (ratio of distance between the tangents at top and bottom contacts to bubble diameter) the deviation between the result obtained for two point forces and the exact solution is only visible in the form of a small indentation at the point contacts. (b) For a large deformation of 20% the model overestimates the lateral dimension of the bubble and leads to large indentations at the point contacts.

5. Force-displacement relation for isotropic compression of a foam

We may use the above equation to derive a force-displacement relation for an isotropically compressed bubble (e.g. in the hexagonal arrangement), as considered by Lacasse et al. [19]. We will here express it in terms of the *osmotic compressibility*, by which we mean the compressibility of a wet foam, allowing the liquid fraction to vary, while the gas is, as assumed above, incompressible. This is related to the concept of an *osmotic pressure* of a foam, which was introduced by Princen [25].

Let us consider the case of a uniform compression of a symmetric structure, such as the hexagonal confinement as shown in Fig. 5, for which Princen gave some analytical results in 1979 [25].

For a single bubble the number of contacts Z can take any integral value; space filling structures require $Z = 3$ (triangular packing), $Z = 4$ (square) or $Z = 6$ (hexagonal) with $Z = 2$ corresponding to a linear bubble chain.

Using the above relationship, Eq. (9), and imposing Z forces of magnitude f , with all radial displacements δr_i set equal to δr we arrive at

$$\delta r = \frac{f}{2\pi} \sum_{n=1}^Z g(n\Delta\theta), \quad (10)$$

where $\Delta\theta = 2\pi/Z$.

Evaluation of the sum results in

$$\delta r = \frac{\pi}{6Z} f, \quad (11)$$

as shown in Appendix C. While in the bubble model the corresponding equation is $\delta r = f$, here the osmotic compressibility varies approximately as the inverse of the contact number Z . More precisely, Eq. (11) is the asymptotic form as Z tends to infinity; it agrees with the result from Lacasse et al. in the limit of small compression [19]. This result is at first surprising, if one is accustomed to thinking in terms of simple pairwise forces. Those described in Section 4 are quite different.

The source of this scaling is the condition of incompressibility of the contents of the bubbles which lies behind the displacement/force relation for *different* contacts. Corresponding rules may be expected to occur more generally in other branches of material science, such as the theory of granular materials, wherever the constituent materials have a relatively low compressibility, as is commonly the case [26].

The example of a uniform compression of a symmetric structure is relatively trivial because the identification of contacts is self-evident, indeed unvarying, for all values of liquid fraction. In general, and particularly for a *disordered* foam, the elimination or creation of contacts (that is, topological changes) present practical difficulties in the implementation of this method in the more general context of typical 2d foams. We will address these elsewhere, in comparison with accurate simulations using the PLAT software [7].

6. Conclusion

The 2d case has fulfilled our expectations of a simple and transparent theory of bubble interactions close to the wet limit. In particular, the origin of the variation of compressibility with contact number, contained in the work of Lacasse et al. [19], is clearly exposed.

In a further paper, initial results of simulations of extended 2d foams, based on the interactions derived above, will be presented and compared in detail with the results of the software PLAT [5-7,27]. These will indicate the range of validity of this formulation, which should extend beyond the immediate vicinity of the wet limit, for many purposes.

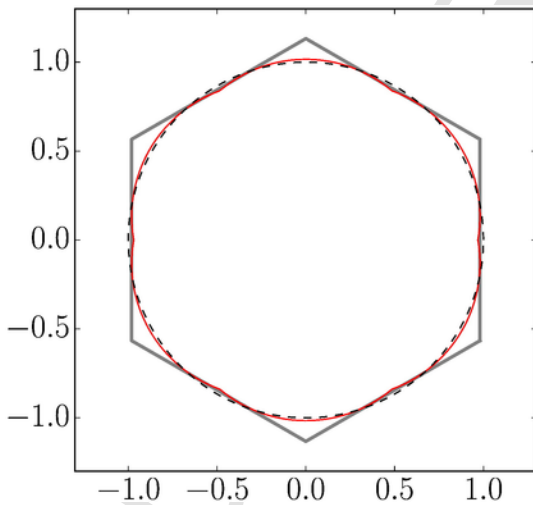


Fig. 5. Example calculations for a bubble confined in a hexagonal box. The dashed line shows the undeformed bubble, the solid line corresponds to the Morse-Witten solution $r(\theta) = 1 + \sum_{j=0}^5 \delta r(\theta + j\pi/3)$.

Modifications to deal with bubble polydispersity will also enable the computation of both shear modulus and yield stress as a function of liquid fraction for disordered foams. Of particular interest will be the variation of the average contact number with liquid fraction, close to the wet limit. While recent extensive PLAT calculations show a linear variation, attributed to bubble deformation [28], in the bubble model the average contact number increases with the square root away from this limit [29].

Interactions in 3d are more subtle, and the subject of a forthcoming review paper [23] whose purpose is partly didactic, as in the present paper.

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Appendix A. Derivation of differential equation for displacement $\delta r(\theta)$, Eq. (2)

The curvature of a curve defined in polar coordinates $R(\theta)$ is given by

$$\kappa(\theta) = \frac{R^2 + 2R_\theta^2 - RR_{\theta\theta}}{(R^2 + R_\theta^2)^{3/2}} \quad (\text{A.1})$$

where $R_\theta = \frac{dR}{d\theta}$ and $R_{\theta\theta} = \frac{d^2R}{d\theta^2}$. Writing $R(\theta) = R_0 + \delta R(\theta)$ and linearising the expression for the curvature leads to

$$\kappa(\theta) = \frac{1}{R_0} - \left(\frac{d^2}{d\theta^2} + 1 \right) \frac{\delta R(\theta)}{R_0^2}. \quad (\text{A.2})$$

A free bubble immersed in water has a uniform curvature, $\kappa = R_0^{-1}$, and its radius is related to the difference between internal and external pressure, p_{i0} and p_{e0} , via the Laplace-Young equation. In two dimensions this is given by $\gamma/R_0 = p_{i0} - p_{e0}$, where γ is the surface tension of a gas-liquid interface.

In the presence of an additional external force acting on the bubble, e.g. due to a contact with a wall or another bubble, the Laplace-Young equation takes the form

$$\gamma/R(\theta) = \gamma\kappa(\theta) = p_i - p_e(\theta), \quad (\text{A.3})$$

where we have adopted the notation $p_i = p_{i0} + \delta p_i$ and $p_e(\theta) = p_{e0} + \delta p_e(\theta)$ from Morse and Witten [13]. Inserting the linearised expression for the curvature, $\kappa(\theta)$ (Eq. (A.2)), then results in

$$-\left(\frac{d^2}{d\theta^2} + 1 \right) \frac{\delta R(\theta)}{R_0} = \frac{R_0}{\gamma} (\delta p_i - \delta p_e(\theta)). \quad (\text{A.4})$$

For the case of a bubble subject to a force F at a single point (at $\theta = 0$) this force will need to be balanced via a uniform body force, arising from an increase in internal pressure. Its gradient is given as $F/(R_0^2\pi)$ (Pascal’s law), resulting in

$\delta p_i - \delta p_e(\theta) = \frac{F}{R_0^2\pi} R(\theta) \cos \theta + A \simeq \frac{F}{R_0^2\pi} \cos \theta + A$, where the integration constant A will later be chosen so as to maintain constant bubble area.

Inserting into the right hand side of Eq. (A.4) then results in

$$-\left(\frac{d^2}{d\theta^2} + 1 \right) \delta r(\theta) = a + \frac{f}{\pi} \cos \theta, \quad (\text{A.5})$$

where we have introduced the dimensionless quantities $\delta r(\theta) = \delta R(\theta)/R_0$, $f = F/\gamma$ and $a = R_0 A/\gamma$. Eq. (A.5) features as Eq. (3) in the main text.

Appendix B. Description of capped shape as a function of force f

We refer to the sketch of Fig. 3 for the definition of cap length l and height h and also for the definition of the angle θ_m .

At the end points of the cap, $r(\theta) \cos(\theta)$ is a maximum with respect to θ . This leads to $\theta_m = f\pi/(2\pi - f)$. The cap length is then given by $l = 2(1 + \delta r(\theta_m)) \sin \theta_m \simeq f(1 - f/(2\pi))$.

$$h = r(\theta_m) \cos \theta_m - r(\theta = 0)$$

The cap height is given by $\simeq (f/2)\theta_m$. The change in area

$$\simeq \frac{f^2}{4} (1 + f/(2\pi))$$

of the enclosed curve is thus of order f^3 .

Appendix C. Derivation of Eq. (11), $\delta r = \frac{\pi}{6Z} f$.

Eq. (10) is of a kind familiar from numerical integration and we will evaluate it by considering the integral $\int_0^{2\pi} g(\theta) d\theta$. This may be approximated using the composite trapezoid rule as

$$\int_0^{2\pi} g(\theta) d\theta = \sum_{n=1}^Z g(n\Delta\theta) \Delta\theta + \frac{1}{12} \sum_{n=1}^Z g''(n\Delta\theta) (\Delta\theta)^3 + \text{higher order}, \quad (\text{C.1})$$

where we took into account that $g(0) = g(2\pi)$.

Since the left hand side vanishes by the condition of incompressibility, see Eq. (6), we arrive at

$$\Delta\theta \sum_{n=1}^Z g(n\Delta\theta) = -\frac{1}{12} (\Delta\theta)^3 \sum_{n=1}^Z g''(n\Delta\theta). \quad (\text{C.2})$$

We can now write the right hand side in turn as an integral, and evaluate this as $\Delta\theta \sum_{n=1}^Z g''(n\Delta\theta) \simeq \int_0^{2\pi} g''(\theta) d\theta = g'(\theta)|_0^{2\pi}$, neglecting higher order terms. For our function $g(\theta)$, Eq. (8), this results in -2π and thus $\Delta\theta \sum_{n=1}^Z g(n\Delta\theta) \simeq \frac{\pi}{6} (\Delta\theta)^2$.

Returning to our original equation, Eq. (10), this then gives $\delta r = \frac{f}{2\pi} \frac{\pi}{6} \Delta\theta$. Inserting $\Delta\theta = 2\pi/Z$ results in $\delta r = f \frac{\pi}{6Z}$ (see Eq. (11)), in leading order in Z^{-1} .

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