Tracing the Connection between Different Expressions for the Laplace Pressure of a General Curved Interface

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We compare the expression for the Laplace pressure across a general curved interface of local mean and Gaussian curvatures, H and K, derived by Boruvka and Neumann, *J. Chem. Phys.* **66**, 5464 (1977), with a corresponding expression given in terms of the mean and deviatoric curvatures, H and D, obtained by Kralchevsky, *J. Colloid Interface Sci.* **137**, 217 (1990). The two expressions are shown to yield exactly the same result when the contribution due to the thermodynamic surface shearing tension, ζ, is negligible. In the case of a general curved interface, the mechanical (σ) and thermodynamical (γ) interfacial tensions are different, and alternative expressions for the Laplace pressure can be based on either of these two quantities. They are compared and discussed for the special case of an interface that obeys the Helfrich bending energy expression. © 1993 Academic Press, Inc.

INTRODUCTION

In the bulk of a bicontinuous Winsor III (middle) microemulsion phase which coexists with excess oil and water phases, there are internal, surfactant-laden oil–water interfaces, seemingly in the form of thermally agitated, periodic minimal surfaces (1, 2). An analogous situation is encountered with L_3 (sponge) phases where (swollen) bilayers separating either oil or water domains are present, likewise shaped similarly as distorted periodic minimal surfaces, and perhaps also in certain bicontinuous cubic liquid crystals. In all of these cases we may hence assume that the equilibrium configuration of the extended internal interface corresponds approximately to an infinite periodic minimal surface. Such an interface is characterized by a mean curvature, $H = \frac{1}{2}(c_1 + c_2)$, that is everywhere equal to zero and a Gaussian curvature, $K = c_1 c_2$, that varies with the curvilinear coordinates along the interface, from some negative minimum value to a maximum value equal to zero at the flat points.

Moreover, it is an important experimental condition for the formation of a fluid structure of this kind that the interfacial tension γ attains some ultralow minimum value, below about 10^{-2} mN m^{-1}, at the spontaneous curvature, H_0. In addition, H_0 must be adjusted to zero (2). Hence, we are dealing with a regime where curvature effects have to be taken into account explicitly.

We may base a theoretical description of a fluid, *thermodynamically open*, interface composed of *soluble* components in a Winsor III or L_3 phase on an *ansatz* as to its superficial excess free energy density, i.e., its thermodynamic (Gibbsian) interfacial tension, γ, of the general type

$$\gamma = \gamma(T, \mu_i, H, K) \qquad [1]$$

or, alternatively,

$$\gamma = \gamma(T, \mu_i, H, D), \qquad [2]$$

where D stands for the *deviatoric* curvature, $\frac{1}{2}(c_1 - c_2)$.

In particular, we note that the Laplace pressure, Δp, across an internal interface in a bicontinuous Winsor III or L_3 phase must vanish at equilibrium, since, otherwise, the physicochemical equilibrium condition, $\mu_i = $ constant, cannot be satisfied throughout the system. As we have discussed earlier (2), an expression for the Laplace pressure results from minimizing the grand Ω-potential of the interfacial system, subject to a constraint of constant chemical potentials. This is in full analogy with the conventional way of deriving the Young–Laplace equation for the case of an interface of uniform curvature.

Toward this background one realizes that we need to have available a generalized expression for the Laplace pressure, Δp, that includes the case of an interface of *nonuniform* curvature. For the less general case of a quasi-uniform (i.e., with slowly varying curvatures) interface, and an *arbitrary dividing surface*, the following expression was derived by Murphy (3) and was given on a slightly different, less transparent, form by Melrose (4)

$$\Delta p = 2H\gamma - C_1(2H^2 - K) - 2C_2 HK, \qquad [3]$$

where $C_1 = (\partial \gamma / \partial H)_{T,\mu_i,K}$ and $C_2 = (\partial \gamma / \partial K)_{T,\mu_i,H}$. It should

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be pointed out that the derivation made by Melrose of this expression for Δp is based on considerations of parallel displacements of the dividing surface which are proper only in the case of interfaces with slowly varying or constant curvature. Hence, it is applicable, e.g., to spherical or cylindrical microemulsion droplets in Winsor type microemulsions where a $\Delta p = 0$ condition is also of fundamental importance (5). Since the correct expression for Δp may be of interest also in other connections we shall discuss the Laplace pressure below in a general manner.

GENERAL EXPRESSIONS FOR THE LAPLACE PRESSURE

For the more general case of an interface of non-uniform curvature, essentially two universal Δp-expressions have been presented in the literature, one due to Boruvka and Neumann (6) in 1977 and one due to Kralchevsky (7) in 1990. Murphy's theory (3), later on quoted by Neogi and Friberg (8), was originally intended to cover also the case of nonuniform curvature but due to a mathematical inadvertency, subsequently pointed out in Ref. (7), it is actually valid only for quasi-uniform surfaces. In addition, Markin et al. (9) stressed that Boruvka and Neumann's definition of the surface tension differs from the Gibbs surface tension that is commonly used. If we account for this purely formal difference, Boruvka and Neumann's expression for the Laplace pressure has the following form

$$\Delta p = 2H\gamma - C_1(2H^2 - K) - 2C_2HK$$
$$- \tfrac{1}{2}\nabla_S^2 C_1 - K\nabla_S^* \cdot (\nabla_S C_2) \quad [4]$$

and is valid for an arbitrary dividing surface. Here ∇_S^2 is the Laplace–Beltrami operator on the surface and $\nabla_S^* = b^{ij}\mathbf{r}_i\nabla_j$ (where $\mathbf{b} = -\nabla_S\mathbf{n}$ is the second fundamental tensor of the surface) is a special operator introduced by Weatherburn (cf. Ref. (10)). The last two terms containing ∇_S and ∇_S^2 of the above Δp-expression drop out, of course, for an interface of quasi-uniform curvature where γ is (approximately) independent of the curvilinear surface coordinates.

The Boruvka and Neumann expression, Eq. [4], encompasses in essence the "vesicle shape equation" derived by Ou-Yang and Helfrich (11) by means of a variational approach for the special case of a geometrically closed bilayer interface obeying the ordinary Helfrich bending energy expression (cf. Ref. (2)):

$$\Delta p = 2H\gamma_0 - 4k_c(H - H_0)(H^2 + HH_0 - K) - 2k_c\nabla_S^2 H$$
$$= 2H\gamma_\infty - 4k_c[H(H^2 - K) + H_0K] - 2k_c\nabla_S^2 H, \quad [5]$$

where $\gamma_0 = \gamma\ (H = H_0, K = 0)$ and where k_c and $\bar{k}_c$ are two (state-dependent) constants relating to variations of the mean and Gaussian curvatures, respectively. The constant γ_∞ de-

notes the interfacial tension of the planar interface.

Now, by invoking the identity

$$K\nabla_S^* = 2H\nabla_S - \mathbf{b}\cdot\nabla_S \quad [6]$$

which results from the definition of the Weatherburn operator (10), ∇_S^*, we obtain the relation

$$K\nabla_S^* \cdot \nabla_S C_2 = 2H\nabla_S^2 C_2 - \mathbf{b} : \nabla_S\nabla_S C_2, \quad [7]$$

where, as above, $\mathbf{b} = -\nabla_S\mathbf{n}$ is the second fundamental tensor of the interface, $\mathbf{n}$ being the surface normal. Hence, the Boruvka and Neumann expression can be rewritten as

$$\Delta p = 2H\gamma - C_1(2H^2 - K) - 2C_2HK$$
$$- \tfrac{1}{2}\nabla_S^2 C_1 - 2H\nabla_S^2 C_2 + \mathbf{b} : \nabla_S\nabla_S C_2. \quad [8]$$

The corresponding Δp-expression derived by Kralchevsky (7) by a variational method, employing H and D as the curvature variables, has the following form

$$\Delta p = 2H\gamma + 2D\zeta - (H^2 + D^2)B$$
$$- 2HD\Theta - \tfrac{1}{2}(a^{\mu\sigma}B + q^{\mu\sigma}\Theta)_{,\mu\sigma} \quad [9]$$

after correction for an unfortunate mistake in Ref. (7) where it was erroneously assumed that the covariant derivatives of the components of the tensor $\mathbf{q}$, i.e., $q_{\lambda\mu,\nu}$, are equal to zero. In this equation, B and Θ are the sum and difference, respectively, of the eigenvalues of the bending moment tensor, cf., Ref. (12). In the case of a surfactant-laden interface in full equilibrium with the solution we have $B = (\partial\gamma/\partial H)_{T,\mu_i,D}$ and $\Theta = (\partial\gamma/\partial D)_{T,\mu_i,H}$. Furthermore, $a^{\mu\sigma}$ are the contravariant components of the first fundamental tensor of the interface whereas the tensor $\mathbf{q}$ is defined in the Appendix. The term $2D\zeta$ will be discussed below.

From the definitions of D, B, and Θ one derives the relations

$$D = \sqrt{H^2 - K} \quad [10]$$

$$B = C_1 + 2C_2H \quad [11]$$

$$\Theta = -2C_2D \quad [12]$$

by means of which we can readily establish that

$$(H^2 + D^2)B + 2HD\Theta = C_1(2H^2 - K) - 2C_2HK. \quad [13]$$

Moreover, in the Appendix it is shown that

$$\tfrac{1}{2}(a^{\mu\nu}B + q^{\mu\nu}\Theta)_{,\mu\nu} = \tfrac{1}{2}\nabla_S^2 C_1 + 2H\nabla_S^2 C_2 - \mathbf{b} : \nabla_S\nabla_S C_2 \quad [14]$$

implying that the above expression given by Eq. [9] for Δp may be written in the following alternative way

$$\Delta p = 2H\gamma + 2D\zeta - C_1(2H^2 - K) - 2C_2HK$$
$$- \tfrac{1}{2}\nabla_S^2 C_1 - 2H\nabla_S^2 C_2 + \mathbf{b} : \nabla_S\nabla_S C_2 \quad [15]$$

which differs from Boruvka and Neumann's expression only by the presence of the additional term $2D\zeta$.

The variable ζ introduced by Gurkov and Kralchevsky (12) is the *thermodynamic shearing tension* and it corresponds physically to the free energy per unit area needed to elastically shear an interface while keeping its extension unchanged. Formally, ζ is related to the *mechanical shearing tension*, η, by means of the equation (12)

$$\zeta = \eta + \tfrac{1}{2}BD + \tfrac{1}{2}\Theta H \quad [16]$$

with η being defined by

$$\eta = \tfrac{1}{2}(\sigma_1 - \sigma_2), \quad [17]$$

where σ_1 and σ_2 are the principal components of the (mechanical) interfacial stress tensor. Moreover, in terms of these components we also define the *mechanical interfacial* tension

$$\sigma = \tfrac{1}{2}(\sigma_1 + \sigma_2) \quad [18]$$

and an equation similar to Eq. [16] serves to relate σ and γ (cf., Ref. (12)):

$$\sigma = \gamma - \tfrac{1}{2}BH - \tfrac{1}{2}\Theta D. \quad [19]$$

Upon combining Eqs. [9], [16] and [19] we obtain

$$\Delta p = 2H\sigma + 2D\eta - \tfrac{1}{2}(a^{\mu\nu}B + q^{\mu\nu}\Theta)_{,\mu\nu} \quad [20]$$

which is an alternative (and more compact) form of the Δp expression, Eq. [9]. As shown in Ref. (13), Eq. [20] can be independently derived in a more direct manner as a normally resolved local surface balance of the linear momentum. Hence, in this way we are able to confirm the validity of Eq. [9] by means of referring to the force balance perpendicular to the interface.

APPLICATION TO HELFRICH INTERFACES

Remaining questions are what values the difference between the mechanical and thermodynamical interfacial tensions, σ and γ, as well as between their shearing counterparts, η and ζ may assume. To elucidate this point, let us consider a partly closed interface in the thermodynamical sense and let us specify its flexural rheology. A frequently employed model of Helfrich (15) implies that the free energy of purely flexural deformation per unit area, w_f, can be written in the form:

$$w_f = 2k_c(H - H_0)^2 + \bar{k}_c K, \quad [21]$$

where, as already noted above, k_c and $\bar{k}_c$ are (state-dependent) coefficients of bending and torsion elasticity and H_0 is the spontaneous *mean* curvature (a spontaneous *deviatoric* curvature is not introduced in this model). For the sake of brevity we shall call each interface for which Eq. [21] holds a *Helfrich interface*. Since $K = H^2 - D^2$, in accordance with Eq. [87] in Ref. (12), one derives keeping T and the appropriate chemical potentials constant, and the state of dilatational and shearing strain fixed:

$$B = (\partial w_f/\partial H)_D = 4k_c(H - H_0) + 2\bar{k}_c H, \quad [22]$$

and

$$\Theta = (\partial w_f/\partial D)_H = -2\bar{k}_c D. \quad [23]$$

Substitution of Eqs. [22] and [23] into Eqs. [19] and [16] now yields

$$\gamma - \sigma = 2k_c H(H - H_0) + \bar{k}_c K, \quad [24]$$

and

$$\zeta - \eta = 2k_c D(H - H_0). \quad [25]$$

In the special case where $\zeta = 0$ we obtain from Eqs. [24] and [25]

$$\sigma_1 = \gamma - 2k_c(H - H_0)(H + D) - \bar{k}_c K \quad [26]$$

$$\sigma_2 = \gamma - 2k_c(H - H_0)(H - D) - \bar{k}_c K \quad [27]$$

showing that the surface stress tensor is in general *anisotropic* even when $\zeta = 0$.

Turning next to a spherical interface with arbitrary ζ and with $D = 0$ and $K = H^2$, Eqs. [24] and [25] reduce to

$$\gamma = \sigma + \tfrac{1}{2}BH = \sigma - 2k_c HH_0 + (2k_c + \bar{k}_c)H^2 \quad [28]$$

and

$$\zeta = \eta, \quad [29]$$

respectively. The first of the above equations, Eq. [28], implies that $\gamma = \sigma$ when $B = 0$, i.e., when employing the surface of tension as the dividing surface, the location of which becomes determined by the condition $H/H_0 = 2k_c/(2k_c + \bar{k}_c)$, whereas Eq. [29] shows that the surface stress tensor is always *isotropic* for a spherical Helfrich interface for which $\zeta = 0$.

For the special case of a *cylindrical* interface ($D = H$, $K = 0$) we derive from Eqs. [24] and [25] that

$$\gamma - \sigma = 2k_c H(H - H_0) = \zeta - \eta. \quad [30]$$

Hence, in contrast to the spherical case one generally has that $\zeta \neq \eta$ for a cylindrical Helfrich interface. It is only when H is equal to the spontaneous curvature, H_0, that we obtain $\gamma = \sigma$ and $\zeta = \eta$. It may also be worth noting that for a cylindrical interface such that $\zeta = 0$ we get from Eqs. [26] and [27]

$$\sigma_1 = \gamma - 4k_c H(H - H_0) \qquad [31]$$

and

$$\sigma_2 = \gamma, \qquad [32]$$

where the subscript 2 refers to the direction along the cylinder axis.

Finally, let us specify the expression for Δp in the case of a general Helfrich interface of arbitrary shape. By using Eqs. [22], [23], [A.2], and Codazzi's equation (see Appendix) one derives the relationship

$$(a^{\mu\nu}B + q^{\mu\nu}\Theta)_{,\nu} = 4k_c H^{,\mu}. \qquad [33]$$

It is noteworthy that Eq. [33] does not contain $\bar{k}_c$. Then by substituting Eq. [33] into Eq. [20] we obtain

$$\Delta p = 2H\sigma + 2D\eta - 2k_c \nabla_S^2 H, \qquad [34]$$

where, in the case of an interface composed entirely of soluble components and such that $\zeta = 0$, $2(H\sigma + D\eta)$ equals the sum of the corresponding terms in Eq. [5].

For capillary waves of small enough amplitude, u, on a flat interface, Eq. [34] can be linearized. One has (17)

$$2H \approx \nabla_S^2 u; \qquad \Delta p = gu\Delta\rho, \qquad [35]$$

where $\Delta\rho$ is the density difference between the two neighboring phases and g is the gravity acceleration. Note that in the linear approximation which is valid insofar as the quadratic terms with respect to the curvature are negligible, we have $\sigma = \gamma$ and $\eta = \zeta$, cf., Eqs. [24] and [25]. Then, for a fluid interface with $\eta = \zeta = 0$, Eq. [34] reduces to

$$\gamma\nabla_S^2 u - k_c\nabla_S^4 u = gu\Delta\rho. \qquad [36]$$

In particular it follows from Eq. [36] that the mean square amplitude of the thermal corrugations with wave number q reads (16, 18)

$$\langle u_q^2 \rangle = k_B T A^{-1}(g\Delta\rho + \gamma q^2 + k_c q^4)^{-1}, \qquad [37]$$

where A is the area of the interface and k_B is the Boltzmann constant. Eq. [37] is widely used to calculate $\gamma = \gamma_\infty$ and k_c from data on light scattering due to capillary waves (16). Our analysis confirms that Eqs. [36] and [37] do not contain

the Gaussian (torsion) elasticity, $\bar{k}_c$. Furthermore, the approximations made above can be shown to be valid for $\gamma_\infty \geqslant 10^{-3}$ N/m.

DISCUSSION AND CONCLUSIONS

1. We have demonstrated above that the two forms of the generalized Laplace equation, Eqs. [4] and [9], derived in Refs. (6) and (7), respectively, are equivalent when the contribution of the shearing tension is negligible ($\zeta = 0$). Moreover, the absence of a ζ-term in Eq. [4] is not indicative of any major disagreement with Eq. [9] since, in effect, Boruvka and Neumann implicitly assumed $\zeta = 0$ from the very beginning of their treatment.

2. For a general curved interface, the mechanical and thermodynamical interfacial tensions are different ($\gamma \neq \sigma$, $\zeta \neq \eta$) and, correspondingly, there are in the main two different but equivalent expressions for the Laplace pressure. One of them, Eq. [9] or Eq. [15], is formulated in terms of γ and ζ, and the other, Eq. [20], in terms of σ and η.

3. Two alternative definitions of a *fluid* interface are conceivable which are based either on the thermodynamic interfacial properties or on the corresponding mechanical properties. According to the former definition, the work of local interfacial shear deformation of a *fluid* interface should be zero, i.e., $\zeta = 0$. However, Eqs. [16], [26] and [27] show that even when $\zeta = 0$, the mechanical shearing tension, η, is not necessarily equal to zero; i.e., the surface stress tensor may be anisotropic for such a fluid interface at quasi-static conditions.

Alternatively, one may define a *fluid* interface by means of a two-dimensional version of Pascal's law, that is, the surface stress tensor is required to be isotropic, hence $\sigma_1 = \sigma_2$ and $\eta = 0$. However, in such a case the work of shear is not always zero but is determined by the bending and torsion moments, i.e., $\zeta = (BD + \Theta H)/2$, cf., Eq. [16]. According to this expression $\zeta = 0$ only for planar and spherical geometries. One realizes that the two alternative definitions of a fluid interface are not equivalent for a general curved interface. The above complications originate from the fact that a *flexural* deformation of a surface is usually coupled with a *shear* deformation and vice versa. Of course, it is largely a matter of convention what is to be called a "fluid" interface. What makes physical sense in the end is whether a real curved boundary between two fluid phases complies with $\zeta = 0$ or $\eta = 0$.

However, there is one special case where $\zeta = 0$ and this is when the three-dimensional pressure tensor, $\mathbf{P}$, is transversely isotropic throughout the interfacial region, i.e., when

$$\mathbf{P} = P_T \mathbf{U}_S + P_n \mathbf{nn} \qquad [38]$$

holds, where $\mathbf{U}_S$ is the surface idemfactor, cf., Ref. (7). It

can then be shown, using the equations of Ref. (7), that

$$\eta = -D \int_{\lambda_1}^{\lambda_2} (P_T - \bar{P}) \lambda \, d\lambda \qquad [39]$$

where λ is the perpendicular distance from the point in space to the dividing surface, i.e., $\lambda = 0$ on the dividing surface. This equation shows in a more detailed manner than Eq. [16] that η is generally different from zero even when $\zeta = 0$, except for certain special cases, e.g., for planar and spherical geometries.

Furthermore, we may note that current (mean field) theories of hydrocarbon chains packed in interfaces and liquid crystals do, in fact, assume the pressure tensor, **P**, to be transversely isotropic (19, 20).

APPENDIX

Here we derive the relation

$$\tfrac{1}{2}(a^{\mu\nu}B + q^{\mu\nu}\Theta)_{,\mu\nu} = \tfrac{1}{2}\nabla_S^2 C_1 + 2H\nabla_S^2 C_2 - \mathbf{b}{:}\nabla_S\nabla_S C_2. \qquad [A.1]$$

In the following we shall denote the left and right sides of this equation by Q_1 and Q_2, respectively.

Using the relation [12] and the identity

$$Dq^{\mu\nu} = b^{\mu\nu} - Ha^{\mu\nu} \qquad [A.2]$$

which may also be written $D\mathbf{q} = \mathbf{b} - H\mathbf{U}_S$, where $\mathbf{U}_S$ is the identity tensor (idemfactor) on the surface, one obtains

$$Q_1 \equiv \tfrac{1}{2}(a^{\mu\nu}B + q^{\mu\nu}\Theta)_{,\mu\nu} = \tfrac{1}{2}a^{\mu\nu}B_{,\mu\nu} + Ha^{\mu\nu}(C_2)_{,\mu\nu}$$
$$- b^{\mu\nu}(C_2)_{,\mu\nu} - 2(C_2)_{,\mu}(Dq^{\mu\nu})_{,\nu} - C_2(Dq^{\mu\nu})_{,\mu\nu}. \qquad [A.3]$$

Using the above expression for $Dq^{\mu\nu}$ and Codazzi's equation, $b^{\mu\nu,\sigma} = b^{\mu\sigma,\nu}$ one derives

$$(Dq^{\mu\nu})_{,\nu} = H^{,\mu}; \quad (Dq^{\mu\nu})_{,\mu\nu} = a^{\mu\nu}H_{,\mu\nu}. \qquad [A.4]$$

Combining Eqs. [A.3] and [A.4], we obtain

$$Q_1 = \tfrac{1}{2}a^{\mu\nu}B_{,\mu\nu} + Ha^{\mu\nu}(C_2)_{,\mu\nu} - b^{\mu\nu}(C_2)_{,\mu\nu}$$
$$- 2(C_2)_{,\mu}H^{,\mu} - C_2 a^{\mu\nu}H_{,\mu\nu}. \qquad [A.5]$$

On the other hand, using Eq. [11] it follows that

$$\tfrac{1}{2}\nabla_S^2 C_1 = \tfrac{1}{2}a^{\mu\nu}(C_1)_{,\mu\nu} = \tfrac{1}{2}a^{\mu\nu}B_{,\mu\nu} - Ha^{\mu\nu}(C_2)_{,\mu\nu}$$
$$- 2(C_2)_{,\mu}H^{,\mu} - C_2 a^{\mu\nu}H_{,\mu\nu}. \qquad [A.6]$$

In addition, one has the following relations

$$2H\nabla_S^2 C_2 = 2Ha^{\mu\nu}(C_2)_{,\mu\nu}; \quad \mathbf{b}:\nabla_S\nabla_S C_2 = b^{\mu\nu}(C_2)_{,\mu\nu}. \qquad [A.7]$$

A combination of the last two equations finally yields

$$Q_2 \equiv \tfrac{1}{2}\nabla_S^2 C_1 + 2H\nabla_S^2 C_2 - \mathbf{b}:\nabla_S\nabla_S C_2 = Q_1 \qquad [A.8]$$

which was to be demonstrated.

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