ESI: Appendices for Conditions under which a natural iterative method for calculating the orientation distribution of rodlike particles decreases the free energy at each step

Alan E. Berger[a]

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Appendix 1: Derivation of the values of w_m for $W(\gamma) = \sin(\gamma)$

We derive the values for $\{w_m\}$ as done in Kayser and Raveché [1] but for the ODF normalization used herein. We are expressing $W(\gamma) = \sin(\gamma)$ as the series $\sum_{m=0}^{\infty} w_m P_m(\cos\gamma)$.

The following known integral involving Legendre polynomials P_m [2, page 798 equation 7.132.1] naturally arises when expanding $W(\gamma) = \sin(\gamma)$ as a linear combination of Legendre polynomials $P_m(\cos\gamma)$. Since this W satisfies $W(\pi - \gamma) = W(\gamma)$, and $P_m(-x) = -P_m(x)$ for m odd, w_m will be 0 for odd integers m, and so we only need the result when $m = 0$ or when $m = 2n$ is an even positive integer:

$$\int_{-1}^{1} (1 - x^2)^{\frac{1}{2}} P_{2n}(x)\, dx = \frac{\pi\Gamma(3/2)\Gamma(3/2)}{\Gamma(n+2)\Gamma((3/2)-n)\Gamma(n+1)\Gamma((1/2)-n)} \tag{S.1}$$

Using the change of variable $x = \cos(\gamma)$ in the integral on the left side of the above, one finds

[a] *Department of Surgery, Johns Hopkins University School of Medicine, Baltimore, MD 21287, USA. E-mail: aberger9@jhmi.edu*

$$\int_{-1}^{1}(1-x^2)^{\frac{1}{2}}\, P_{2n}(x)\,dx = \int_{0}^{\pi}\sin^2(\gamma)P_{2n}(\cos\gamma)\,d\gamma \tag{S.2}$$

and one then also has the right side of equation (S.1) from page 338 equation 8.14.16 in [3], or page 172, equation (27) in [4]. Note in some sources (other edition(s) of [2], page 316 equation (16) in [5]), there is a typographical error in this formula, having (in error) $\Gamma(n+(5/2))$ instead of the correct $\Gamma(n+2)$. One can quickly check that the incorrect version can not be valid from the case $n=0$, since $\int_0^\pi \sin^2(\gamma)\,d\gamma = \pi/2$.

For $W(\gamma)=\sin(\gamma)$ we have

$$W(\gamma) = \sum_{m=0,2}^{\infty} w_m P_m(\cos\gamma)$$

where the notation means sum over $m=0$ and positive even integers. Let $k=0$ or let $k=2n$ be an even positive integer, and recall that

$$\frac{1}{2}\int_{\gamma=0}^{\pi} P_m(\cos\gamma)P_n(\cos\gamma)\sin(\gamma)\,d\gamma = \frac{1}{2}\int_{-1}^{1} P_m(x)P_n(x)\,dx = \delta_{mn}\frac{1}{(2n+1)} \tag{S.3}$$

where δ_{mn} is 1 when $m=n$ and 0 otherwise (see, for example, Chapter 11 of Weber and Arfken [6]). Then

$$\int_{\gamma=0}^{\pi} W(\gamma)P_k(\cos\gamma)\sin(\gamma)\,d\gamma = \int_{\gamma=0}^{\pi} w_k {P_k}^2(\cos\gamma)\sin(\gamma)\,d\gamma = \frac{2w_k}{2k+1} \tag{S.4}$$

However, the left hand side of the equation above is (for $W(\gamma)=\sin(\gamma)$):

$$\int_{\gamma=0}^{\pi}\sin^2(\gamma)P_k(\cos\gamma)\,d\gamma \tag{S.5}$$

so from equations (S.2) and (S.1), we have (with $k=2n$):

$$\frac{2w_k}{2k+1} = \frac{\pi\Gamma(3/2)\Gamma(3/2)}{\Gamma(n+2)\Gamma((3/2)-n)\Gamma(n+1)\Gamma((1/2)-n)} \tag{S.6}$$

It will be convenient for the rest of this subsection to define

$$v_n = w_k \text{ which is } w_{2n} \tag{S.7}$$

Then we have

$$v_n = w_{2n} = \frac{4n+1}{2} \frac{\pi\Gamma(3/2)\Gamma(3/2)}{\Gamma(n+2)\Gamma((3/2)-n)\Gamma(n+1)\Gamma((1/2)-n)} \tag{S.8}$$

To display the first several values of $v_n = w_{2n}$ we need some values of the Γ function, available from its basic properties, e.g., [3], [6]:

$$\Gamma(1/2) = \pi^{1/2}, \quad \Gamma(3/2) = \pi^{1/2}/2, \quad \Gamma(-1/2) = -2\pi^{1/2}, \quad \Gamma(-3/2) = (4/3)\pi^{1/2} \tag{S.9}$$

Using these in equation (S.8), we find, as in the Appendix of [7] but with adjusted notation:

$$v_0 = \pi/4, \quad v_1 = -5\pi/32, \quad v_2 = -9\pi/256, \quad v_3 = -65\pi/2^{12} \tag{S.10}$$

Using $\Gamma(z+1) = z\,\Gamma(z)$ and so also $\Gamma(y-1) = \Gamma(y)/(y-1)$ with equation (S.8), we have for $n \geq 2$ (cf. the Appendix in [7] but note the different notation, m there is $2n$ here):

$$v_{n+1} = v_n(16n^3 + 20n^2 - 4n - 5)/(16n^3 + 52n^2 + 44n + 8) \tag{S.11}$$

which shows that all of the w_{2n} are negative for $n > 0$. We can use Raabe's test to show that the series $\sum_{n=0}^{\infty} v_n$ is absolutely convergent, since

$$\rho \equiv \lim_{n\to\infty} n(\frac{v_n}{v_{n+1}} - 1) \text{ is } 2$$

(which is > 1 so sufficient for convergence).

Note using equations (S.10) and (S.11) one can verify that the eigenvalues λ_{2n} in equation (2.12) of Kayser and Raveché [1] based on their scaling convention are equal to $8v_n/[\pi(4n+1)]$.

Appendix 2: Derivation of the expansion of $K(\theta_1, \theta_2)$ in terms of Legendre polynomials

We provide this expansion following Kayser and Raveché [1], but for our normalizations. The situation here is a particular case of integral operators with symmetric kernels [8]. A special result for Legendre polynomials made use of in [1], [7] and here is the *addition formula* due to Laplace, available in [9], or page 1274 equation 10.3.38 in [10], or page 1015 addition theorem 8.814 in [2]:

With γ given by $\operatorname{Arc}\cos[\sin(\theta_1)\sin(\theta_2)\cos(\phi)+\cos(\theta_1)\cos(\theta_2)]$, and $n>0$ (S.12)

one has

$$P_n(\cos\gamma) = P_n(\cos\theta_1)P_n(\cos\theta_2) + 2\sum_{m=1}^{n}\frac{(n-m)!}{(n+m)!}P_n^m(\cos\theta_1)P_n^m(\cos\theta_2)\cos(m\phi) \quad \text{(S.13)}$$

where the P_n^m are *associated Legendre polynomials* and recall $P_0(x)\equiv 1$. Now, for the expansion we want, recall that:

$$K(\theta_1,\theta_2) = \frac{1}{2\pi}\int_{\phi=0}^{2\pi} W(\gamma = \operatorname{Arc}\cos[\sin(\theta_1)\sin(\theta_2)\cos(\phi)+\cos(\theta_1)\cos(\theta_2)])\,d\phi \quad \text{(S.14)}$$

We are expressing $W(\gamma)$ as a series in $P_m(\cos\gamma)$:

$$W(\gamma) = \sum_{m=0}^{\infty} w_m P_m(\cos\gamma) \quad \text{(S.15)}$$

Substituting equation (S.15) with γ as in equation (S.12) into equation (S.14) and using the addition formula equation (S.13), we see that the integral over ϕ "removes" the associated Legendre polynomials P_n^m and we have

$$K(\theta_1,\theta_2) = \sum_{m=0}^{\infty} w_m P_m(\cos\theta_1)P_m(\cos\theta_2) \quad \text{(S.16)}$$

Now if $\mathscr{B}(f)$ is defined to be the last term on the right in

$$\begin{aligned} F(f(\theta)) = \\ & A + \frac{1}{2}\int_{\theta=0}^{\pi} f(\theta)\ln f(\theta)\sin(\theta)\,d\theta \;+ \frac{1}{2}\int_{\theta=0}^{\pi} f(\theta)V(\theta)\sin(\theta)\,d\theta \;+ \\ & \frac{B}{2}\int_{\theta_1=0}^{\pi}\int_{\theta_2=0}^{\pi}\frac{1}{4}f(\theta_1)\sin(\theta_1)K(\theta_1,\theta_2)f(\theta_2)\sin(\theta_2)\,d\theta_2\,d\theta_1 \end{aligned} \quad \text{(S.17)}$$

so

$$\mathscr{B}(f) = \frac{B}{2}\frac{1}{2}\int_{\theta_1=0}^{\pi} \frac{1}{2}\int_{\theta_2=0}^{\pi} f(\theta_1)\sin(\theta_1)K(\theta_1,\theta_2)f(\theta_2)\sin(\theta_2)\,d\theta_2\,d\theta_1 \tag{S.18}$$

and we substitute f expanded in terms of Legendre polynomials,

$$f(\theta) = \sum_{n=0}^{\infty}(2n+1)\eta_n P_n(\cos\theta) \quad \text{with } \eta_n = \frac{1}{2}\int_{\theta=0}^{\pi} f(\theta)P_n(\cos\theta)\sin(\theta)\,d\theta \tag{S.19}$$

and K given by equation (S.16) into equation (S.18), and recall equation (S.3), we are left with (cf. equation (9) in [7]):

$$\mathscr{B}(f) = \frac{B}{2}\sum_{m=0}^{\infty} w_m {\eta_m}^2 \tag{S.20}$$

Appendix 3: Justifying the expansion of $f(\theta)$ as a series in $\{P_n(\cos\theta)\}$

We could appeal to Sturm–Liouville theory on $L^2([0,\pi])$ with the weighted measure $d\mu = \sin(\theta)\,d\theta$, as in, for example, Al-Gwaiz [11], but here a direct approach suffices. Recalling that

$$\frac{1}{2}\int_{\theta=0}^{\pi} P_m(\cos\theta)P_n(\cos\theta)\sin(\theta)\,d\theta = \frac{1}{2}\int_{-1}^{1} P_m(x)P_n(x)\,dx = \delta_{mn}\frac{1}{(2n+1)}$$

define the ortho*normal* Legendre polynomials by

$$\mathscr{P}_n(x) = [(2n+1)/2]^{1/2}P_n(x) \tag{S.21}$$

Since the Legendre polynomials $\{P_n(x)\}$ span the polynomials on $[-1,1]$, e.g., pages 506 - 507 in [6], and polynomials are dense in the space of continuous functions $C([-1,1])$ with the maximum norm by the Stone-Weierstrass theorem, e.g., page 159 in [12], and continuous functions are dense in the space of square integrable functions $L^2([-1,1])$, e.g., page 71 in [13] (with the L^2 norm $\|f_2 - f_1\|_2$ defined to be $(\int_{-1}^{1}(f_2(x) - f_1(x))^2\,dx)^{1/2}$), then for any continuous (or L^2) function $\tilde{f}(x)$ on $[-1,1]$, there are unique constants $\{\xi_j\}$ such that

$$\int_{-1}^{1} \left[\tilde{f}(x) - \sum_{j=0}^{J} \xi_j \mathscr{P}_j(x)\right]^2 dx \to 0 \quad \text{as } J \to \infty \tag{S.22}$$

and also

$$\sum_{j=0}^{\infty} {\xi_j}^2 = \int_{-1}^{1} \tilde{f}^2(x)\, dx \tag{S.23}$$

L^2 with its associated norm is the "natural" function space / setting in which to consider expansions in terms of Legendre polynomials (and many other special functions, including classical Fourier series).

Now if $f(\theta)$ is a continuous function on $[0, \pi]$, define $\tilde{f}(x) = f(\text{Arc}\cos(x))$, and let $\{\xi_j\}$ be as in the two equations above. Then using the change of variable $x = T(\theta) \equiv \cos(\theta)$,

$$\int_{-1}^{1} \left[\tilde{f}(x) - \sum_{j=0}^{J} \xi_j \mathscr{P}_j(x)\right]^2 dx = \int_{0}^{\pi} \left[f(\theta) - \sum_{j=0}^{J} \xi_j \mathscr{P}_j(\cos\theta)\right]^2 \sin(\theta)\, d\theta \to 0 \quad \text{as } J \to \infty \tag{S.24}$$

so $f(\theta)$ admits a unique expansion in terms of $\{\mathscr{P}_n(\cos\theta)\}$ and therefore also in terms of $\{P_n(\cos\theta)\}$; and note when $f(\pi - \theta) = f(\theta)$ and so $\tilde{f}(-x) = \tilde{f}(x)$, the coefficients for the odd indexed Legendre polynomials vanish, and similarly for $W(\gamma) = \sin(\gamma)$.

From equation (S.21) and $\{\mathscr{P}_n(x)\}$ being orthonormal, we have

$$\eta_m = \frac{1}{2} \int_{\theta=0}^{\pi} f(\theta) P_m(\cos\theta) \sin(\theta)\, d\theta = \left[\frac{2}{(2m+1)}\right]^{1/2} \frac{1}{2} \int_{-1}^{1} \tilde{f}(x) \mathscr{P}_m(x)\, dx = \left[\frac{2}{(2m+1)}\right]^{1/2} \frac{1}{2} \xi_m \tag{S.25}$$

and hence $\sum_{m=0}^{\infty} {\eta_m}^2$ is finite and $\sum_{m=0,2}^{\infty} w_m {\eta_m}^2$ would be bounded even if all we knew was that $\{w_m\}$ was bounded.

Appendix 4: The "hat" function construct used in proving a standard result in the calculus of variations

That

$$\ln f(\theta_1) = -\frac{B}{2}\int_{\theta_2=0}^{\pi} K(\theta_1,\theta_2) f(\theta_2)\sin(\theta_2)\, d\theta_2 - V(\theta_1) - \lambda \tag{S.26}$$

where the constant λ corresponds to the *Lagrange multiplier* for the normalization constraint $\frac{1}{2}\int_{\theta_1=0}^{\pi} f(\theta_1)\sin(\theta_1)\, d\theta_1 = 1$ follows from

$$\frac{1}{2}\int_{\theta_1=0}^{\pi} g(\theta_1)\left[\ln f(\theta_1) + V(\theta_1) + \frac{B}{2}\int_{\theta_2=0}^{\pi} K(\theta_1,\theta_2) f(\theta_2)\sin(\theta_2)\, d\theta_2\right]\sin(\theta_1)\, d\theta_1$$

being 0 for any continuous function $g(\theta)$ on $[0,\pi]$ for which $\int_0^\pi g(\theta)\sin(\theta)\, d\theta = 0$ and $\max|g(\theta)|$ is sufficiently small is a consequence of the following lemma. The standard "hat" function construct used in the proof is helpful for the discussion in Section 3.4.2

Suppose $v(x)$ is a continuous function on $[0,\pi]$, and for all functions $g(\theta)$ which are continuous on $[0,\pi]$ and satisfy $\int_0^\pi g(\theta)\sin(\theta)\, d\theta = 0$ and, for some fixed positive constant k, $\max|g| \leq k$, it is true that

$$\int_{\theta=0}^{\pi} g(\theta)v(\theta)\sin(\theta)\, d\theta = 0 \tag{S.27}$$

Then $v(x)$ equals a constant on $[0,\pi]$.

This follows since otherwise, by continuity, there are two values $0 < \theta_1 \neq \theta_2 < \pi$ with $v(\theta_1) < v(\theta_2)$. We can then construct a function g for which equation (S.27) fails. Define the "hat" (or "tent" or "spike" or "upside-down" V) function $H(\theta;\theta_c,h,\epsilon)$ to be the piecewise linear function which is 0 at $\theta = \theta_c - h$; ϵ at $\theta = \theta_c$; and 0 at $\theta = \theta_c + h$; and 0 outside of $[\theta - h, \theta + h]$, so the *support* of H is $[\theta - h, \theta + h]$. If we take ϵ and h sufficiently small and define g by

$$g(\theta) = H(\theta;\theta_2,h,\epsilon)/\sin(\theta) - H(\theta;\theta_1,h,\epsilon)/\sin(\theta)$$

then 0 and π will be outside of the support of g so there will be no issue with dividing by $\sin(\theta)$, and $\int g(\theta)v(\theta)\sin(\theta)\, d\theta$ will not be 0, while g satisfies $\int_0^\pi g(\theta)\sin(\theta)\, d\theta = 0$, and $\max|g| \leq k$.

If the condition $\int_0^\pi g(\theta)\sin(\theta)\, d\theta = 0$ were not required, then v would have to be 0 and only one hat function would needed for the proof.

Appendix 5: Locations of bifurcations from the isotropic ODF for general W

When $V = 0$, the isotropic ODF, $f = 1$, is always a solution to the calculus of variations equation (S.26). Here it is shown that potential locations of bifurcation points for (S.26) can readily be found in terms of the coefficients w_m in the expansion of $W(\gamma)$ when $V = 0$ and ODFs are assumed to be cylindrically symmetric.

Given

$$f(\theta) = \eta_0 + \sum_{m=1}^{\infty}(2m+1)\eta_m P_m(\cos\theta) \text{ where } \eta_0 = 1 \tag{S.28}$$

$$W(\gamma) = \sum_{n=0}^{\infty} w_n P_n(\cos\gamma) \text{ with } w_n \leq 0 \text{ for } n > 0, \text{ and } \sum_{n=0}^{\infty} |w_n| \text{ finite.} \tag{S.29}$$

and

$$V(\theta) = \sum_{r=0}^{\infty} v_r P_r(\cos\theta) \tag{S.30}$$

and using the orthogonality of the Legendre polynomials

$$\frac{1}{2}\int_{\theta=0}^{\pi} P_m(\cos\theta)P_n(\cos\theta)\sin(\theta)\,d\theta = \frac{1}{2}\int_{-1}^{1} P_m(x)P_n(x)\,dx = \delta_{mn}\frac{1}{(2n+1)} \tag{S.31}$$

and the expression for the particle interaction contribution to the free energy developed in Appendix 2, the free energy F can be written as

$$F(f(\theta)) = A + \frac{1}{2}\int_{\theta=0}^{\pi} f(\theta)\ln f(\theta)\sin(\theta)\,d\theta \ + \sum_{r=0}^{\infty} v_r\eta_r + \frac{B}{2}w_0 + \frac{B}{2}\sum_{m=1}^{\infty} w_m{\eta_m}^2 \tag{S.32}$$

A necessary condition for f to be a local minimum of F is that, for each $k > 0$, the partial derivative of F with respect to η_k

$$\frac{\partial F(f(\theta))}{\partial \eta_k} = \frac{1}{2}\int_{\theta=0}^{\pi}[(1+\ln f(\theta))(2k+1)P_k(\cos\theta)]\sin(\theta)\,d\theta \ + v_k + Bw_k\eta_k \tag{S.33}$$

is 0, which is equivalent to f in equation (S.28) being a solution of the calculus of variations equation (S.26). When $f = 1$ the first term in the integral above is 0 from equation (S.31) with $m = 0$. The last term is also 0 for the isotropic ODF. However, the middle term is only 0 for all positive k when V is a constant. Restricting ourselves to that case, and recalling (S.31), the only second partial derivatives that could be nonzero at the isotropic ODF are the diagonal ones:

$$\frac{\partial^2 F(f(\theta))}{\partial {\eta_k}^2} = \frac{1}{2}\int_{\theta=0}^{\pi} (1/f(\theta))(2k+1)^2 {P_k}^2(\cos\theta)\sin(\theta)\, d\theta \ + Bw_k \tag{S.34}$$

At the isotropic ODF,

$$\frac{\partial^2 F(f(\theta))}{\partial {\eta_k}^2} = (2k+1) + Bw_k \tag{S.35}$$

If for a given B, this is positive for all $k > 0$, then the isotropic ODF will be a local minimum.

By inspection of (S.35), since k and B are positive, only negative w_k can contribute to instability of the isotropic state. For a given positive integer k with $w_k < 0$,

$$B_k = (2k+1)/(-w_k) \tag{S.36}$$

is the value of B at which (S.35) transitions from positive to negative as B increases. The smallest B_k, denoted by B^*, is the location beyond which the isotropic ODF is no longer a local minimum of F and at which one would in general expect to see a branch of anisotropic local minima. Although each B_k is a value at which there may be a branch of non-isotropic solutions of the calculus of variations equation (S.26), bifurcations from values of $B_k > B^*$ generally do not give rise to local minima of F. Moreover, since the series for W is assumed to be absolutely convergent, $|w_k|$ must go to 0 with increasing k, so there can only be a finite number of k for which B_k is equal to a given B_j. If this number is odd, then there will be a branch of anisotropic solutions of (S.26) at B_j Vollmer [14], Rabinowitz [15].

From the above, for the *Onsager kernel* $W(\gamma) = \sin(\gamma)$ for which the odd index w_k are 0 and $w_2 = -5\pi/32$ (see Appendix 1), B^* will be $B_2 = 32/\pi$. For the *dipolar kernel* $W(\gamma) = -\cos(\gamma) = -P_1(\cos\gamma)$ for which $w_1 = -1$, the only B_k is $B_1 = 3$. For the *Maier–Saupe kernel* $W(\gamma) = \frac{1}{3} - \cos^2(\gamma) = -\frac{2}{3}P_2(\cos\gamma)$ for which w_2 = -2/3, the only B_k is $B_2 = 15/2$. These bifurcation locations agree, as expected, with those found in previous work (after accounting for differences in normalization and notation) [1], [14], [16], [17].

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