Bulk properties of composite media. I. Simplification of bounds on the shear modulus of suspensions of impenetrable spheres

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We study third-order upper and lower bounds on the shear modulus of a model composite made up of equisized, impenetrable spherical inclusions randomly distributed throughout a matrix phase. We determine greatly simplified expressions for the two key multidimensional cluster integrals (involving the three-point distribution function for one of the phases) arising in these bounds. These expressions are obtained by expanding the orientation-dependent terms in the integrand in spherical harmonics and employing the orthogonality property of this basis set. The resulting simplified integrals are in a form that makes them much easier to compute. The approach described here is quite general in the sense that it has application in cases where the spheres are permeable to one another (models of consolidated media such as sandstones and sintered materials) and to the determination of other bulk properties, such as the bulk modulus, thermal/electrical conductivity, and fluid permeability.

I. INTRODUCTION

The problem we are generally concerned with is the theoretical prediction of the effective properties (transport, elastic, electromagnetic, etc.) of disordered composite media. This problem is of considerable fundamental and practical interest[1–7] and is exactly soluble, given the phase properties and the infinite set of n-point correlation functions[8–10] that statistically characterize the composite medium. The complete set of statistical functions is almost never known in practice, however. Under such circumstances one can either opt for some sort of approximate self-consistent scheme[11–13] or methods that enable us to place bounds on the effective property. Both of these methods have their own advantages and disadvantages and have been discussed elsewhere.[5,6]

We shall focus our attention on rigorous bounding techniques, since they provide a means of estimating the bulk property, given limited microstructural information on the heterogeneous material. Rigorous bounds are useful because (1) they enable one to test the merits of a theory; (2) one of the bounds can typically provide a relatively accurate estimate of the property[7]; and (3) as successively more microstructural information is included, the bounds become progressively tighter. The specific problem of interest in the present study is the determination of bounds (described below) on the effective shear modulus (G_e) of a suspension of impenetrable equisized spherical inclusions.

Bounds on the effective elastic moduli that depend upon the n-point probability function S_n of the medium have been derived.[14–17] The $S_n(\mathbf{x}^n)$ ($\mathbf{x}^n \equiv \mathbf{x}_1, \mathbf{x}_2, ..., \mathbf{x}_n$) give the probability of finding n points at positions $\mathbf{x}^n$ all in one of the phases, for example, phase 2. For statistically homogeneous media S_1 is simply equal to the volume fraction of phase 2, ϕ_2. The second-order bounds on the elastic moduli of Hashin and Shtrikman[14] depend on S_1 and, in a trivial way, on S_2. McCoy[15] obtained sharper third-order bounds (that were later simplified by Milton[16]), which involve information about S_1, S_2, and S_3. Subsequently, Milton and Phan-Thien[17] (MPT) derived third-order bounds, which improve upon the McCoy bounds, and new fourth-order bounds. Practical application of third- and fourth-order bounds on the effective elastic moduli has been very slow because of the difficulty involved in ascertaining S_3 and S_4 either theoretically or experimentally.

Employing the formalism of Torquato and Stell[9,10] to systematically represent and compute the S_n, third-order bounds on the effective elastic moduli have been recently computed for suspensions of fully penetrable spheres[18] and for dilute dispersions of spheres distributed with an arbitrary degree of penetrability.[19] More recently, third-order bounds on the effective bulk modulus of a composite with impenetrable spherical inclusions have been calculated.[20]

In this paper we consider the evaluation of the third-order McCoy and MPT bounds on the effective shear modulus G_e of a random distribution of impenetrable equisized spheres in a matrix. In Sec. II we present the MPT bounds and some relevant discussions. In Sec. III we invoke the diagrammatic notation[9] for the terms involved in S_3 and present the two key integrals that operate on these terms. The complicated multidimensional cluster integrals are then simplified as far as possible by using expansions of the orientation-dependent quantities in spherical harmonics and the orthogonality of this basis set. The final result presented at the end of Sec. III is still nontrivial, but in a form that is now tractable on a computer. The computation, which is under progress, will be the subject of a subsequent paper. In passing we may reiterate the fact that the method of attack presented here is quite general in the sense that it can be (and has been) readily and systematically applied to cases in which spheres are permeable to one another[18] and to the determination of other bulk properties, such as the conductivity and bulk modulus of composites,[20] and fluid permeability of porous media.[21] Finally, in Sec. IV we present our conclusions.

II. BOUNDS

For any two-phase isotropic composite, McCoy[15] has derived bounds on the effective shear modulus G_e, given the shear moduli G_1, G_2, and the bulk moduli K_1 and K_2 of the respective phases, the volume fraction of one of the phases (say $\phi_2 = 1 - \phi_1$), and several integrals involving derivatives of certain three-point correlation functions. Milton[16] later showed that the third-order McCoy bounds can be written in terms of ϕ_2 and only two multifold integrals ζ_2 and η_2 (defined below) involving the three-point probability function S_3. Subsequently, MPT[17] derived improved third-order bounds on G_e that also depend upon ϕ_2, ζ_2, and η_2. For conciseness we present here only the MPT bounds, which read

$$\left(\langle G\rangle - \frac{6\phi_1\phi_2(G_1-G_2)^2}{6\langle\tilde{G}\rangle + \Xi^{-1}}\right) \leqslant G_e \leqslant \left(\langle G\rangle - \frac{6\phi_1\phi_2(G_1-G_2)^2}{6\langle\tilde{G}\rangle + \theta}\right), \tag{1}$$

where

$$\Xi = \frac{5\langle 1/G\rangle_\zeta\langle 6/K - 1/G\rangle_\zeta + \langle 1/G\rangle_\eta\langle 2/K + 21/G\rangle_\zeta}{\langle 128/K + 99/G\rangle_\zeta + 45\langle 1/G\rangle_\eta}, \tag{2}$$

$$\theta = \frac{3\langle G\rangle_\eta\langle 6K + 7G\rangle_\zeta - 5\langle G\rangle_\zeta^2}{\langle 2K - G\rangle_\zeta + 5\langle G\rangle_\eta}, \tag{3}$$

and where the angular brackets denote the averages of the following types for any property b:

$$\langle b\rangle = b_1\phi_1 + b_2\phi_2,$$
$$\langle b\rangle_\zeta = b_1\zeta_1 + b_2\zeta_2,$$
$$\langle b\rangle_\eta = b_1\eta_1 + b_2\eta_2,$$
$$\langle\tilde{b}\rangle = b_1\phi_2 + b_2\phi_1. \tag{4}$$

Here the quantities ζ_i and η_i ($i = 1,2$) are the integrals (defined below) over the three-point probability function S_3. To be specific, S_n denotes the probability of finding n points in the particulate phase (here the impenetrable spheres). Then we have

$$\zeta_2 = 1 - \zeta_1 = (9/2\phi_1\phi_2)I[\hat{S}_3], \tag{5}$$

$$\eta_2 = 1 - \eta_1 = \tfrac{5}{21}\zeta_2 + (150/7\phi_1\phi_2)J[\hat{S}_3], \tag{6}$$

and

$$\hat{S}_3(r,s,t) = S_3(r,s,t) - S_2(r)S_2(s)/S_1, \tag{7}$$

where the integral operators I and J are defined as

$$I[f] = \frac{1}{8\pi^2}\int d\mathbf{r}_2\,d\mathbf{r}_3\,f(r_{12},r_{13},r_{23})\,\frac{P_2(\hat{\mathbf{r}}_{12}\cdot\hat{\mathbf{r}}_{13})}{r_{12}^3 r_{13}^3}, \tag{8}$$

and

$$J[f] = \frac{1}{8\pi^2}\int d\mathbf{r}_2\,d\mathbf{r}_3\,f(r_{12},r_{13},r_{23})\,\frac{P_4(\hat{\mathbf{r}}_{12}\cdot\hat{\mathbf{r}}_{13})}{r_{12}^3 r_{13}^3}, \tag{9}$$

P_l being the Legendre polynomial of degree l. It may be mentioned that we operate the I and J on $\hat{S}_3$ defined in (7) instead of S_3 because it ensures the absolute convergence of these integrals.

It may further be noted that ζ_i ($i = 1$ or 2) is the only parameter needed for the third-order bounds on electrical conductivity or bulk modulus. The calculation of this parameter, and hence the evaluation of $I[\hat{S}_3]$, has already been considered in great detail in two previous papers.[20] The parameter ζ_2 is thus well tabulated for our needs, and hence we concern ourselves with the evaluation of the integral $J[\hat{S}_3]$ or the parameter η_2 only. The reader interested in the details of the ζ_2 calculation is referred to the above-mentioned papers.[20]

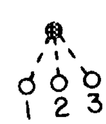
The evaluation of the bounds (1) becomes quite difficult partly because the three-point function S_3 was not available theoretically or experimentally until recently and partly because of the complexity of the integral J [Eq. (9)]. Thus, to our knowledge, this is the first paper to deal with those bounds for the effective shear modulus for the case of a dispersion of equisized impenetrable spheres at arbitrary concentration in a matrix phase.

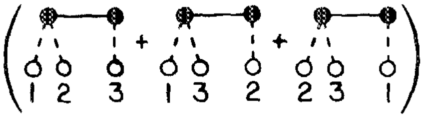
III. SIMPLIFICATION OF $J[\hat{S}_3]$ FOR DISPERSIONS OF IMPENETRABLE SPHERES

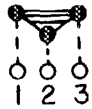
Torquato and Stell[9] have shown that the n-point probability function for the *matrix* phase of dispersions of impenetrable spheres reduces to a finite series expansion in the density ρ, ending at the nth-order term. For our purposes we will use the notation of Ref. 9, except for the fact that S_n now will describe the n-point probability function for the *particle* phase, i.e., the quantity denoted by S_n in the Introduction.[22] As shown by Torquato and Stell,[8] given the n-point probability functions $(S_1, S_2, \ldots, S_n)$ for one of the two phases, one can get any other n-point probability function and, in particular, the n-point probability function for the other phase. Thus, taking the expressions for the one-, two-, and three-point matrix probability functions,[9] we can write the three-point probability function in the particulate phase as

$$S_3 = S_3^{(1)}\phi_2 + S_3^{(2)}\phi_2^2 + S_3^{(3)}\phi_2^3, \tag{10}$$

where $\phi_2 = \rho V_1$ is the particle volume fraction, $V_1 = 4\pi a^3/3$ is the spherical volume of one inclusion, a is the radius of the sphere, and $S_3^{(i)}$ stand for the following diagrams:

$$S_3^{(1)} = \frac{1}{V_1}$$  , (11a)

$$S_3^{(2)} = \frac{1}{V_1^2}\left(\right.$$  $$\left.\right), \quad (11b)$$

$$S_3^{(3)} = \frac{1}{V_1^3}$$  $$. \quad (11c)$$

Here the solid circles stand for dummy position vectors (of some spheres) that are to be integrated over the entire infinite volume,[23] the labeled open circles represent the position vectors $\mathbf{r}_1$, $\mathbf{r}_2$, and $\mathbf{r}_3$ appearing in S_3, the broken line represents the bond

$$m(r) = \begin{cases} 1, & r<a, \\ 0, & r>a, \end{cases} \quad (12)$$

between the two positions involved, the solid line stands for the pair distribution function $g_2 \equiv g$ of the spheres, and the crosshatched triangle for their triplet distribution function g_3.

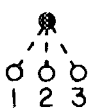
For the calculation of shear modulus, we have to evaluate the functional (9) [evaluation of the other functional (8) has already been done[20]], which may be rewritten as

$$J[f] = \int_0^\infty \frac{dr_{12}}{r_{12}} \int_0^\infty \frac{dr_{13}}{r_{13}} \int_{-1}^{1} d(\cos\theta_{213}) \times P_4(\cos\theta_{213}) f(r_{12},r_{13},r_{23}) . \quad (13)$$

We have to evaluate the above for each of the diagrams in (11). P_4 is the Legendre polynomial of degree 4, and $\cos\theta_{jik} \equiv (\hat{\mathbf{r}}_{ij}\cdot\hat{\mathbf{r}}_{ik})$.

A. Evaluation of $J[S_3^{(1)}]$

The only diagram of (11a) may be evaluated by fixing the origin of coordinates at $\mathbf{r}_1$ and aligning the z axis along $\hat{\mathbf{r}}_{12}$, as follows:



$$\equiv \int d\mathbf{r}_4\, m(r_{14}) m(r_{24}) m(r_{34})$$
$$= \int_0^\infty dr_{14}\, r_{14}^2\, m(r_{14}) \int d\omega_{214}\, m(r_{24}) m(r_{34}) , \quad (14)$$

where $d\omega_{214} \equiv d(\cos\theta_{214}) d\phi$. Following Barker and Monaghan,[24] we expand the angle-dependent functions in Legendre polynomials (more generally, in spherical harmonics). Thus, for example, for $m(r_{24})$ we write

$$m(r_{24}) = m[(r_{12}^2 + r_{14}^2 - 2r_{12}r_{14}\cos\theta_{214})^{1/2}]$$
$$= \sum_{l=0}^{\infty} M_l(r_{12},r_{14}) P_l(\cos\theta_{214}) , \quad (15)$$

where the expansion coefficients are given by (see Appendix A)

$$M_l(r_{12},r_{14}) = \frac{2l+1}{2\pi^2} \int_0^\infty dk\, k^2 \tilde{m}(k) j_l(kr_{12}) j_l(kr_{14}) . \quad (16)$$

$\tilde{m}(k)$ is the Fourier transform of $m(r)$, and $j_l(x)$ is the spherical Bessel function of order l. For the Fourier transform $\tilde{f}(k)$ of a function $f(r)$, we will always use the definition

$$\tilde{f}(k) = \int d\mathbf{r}\, f(r) \exp(i\mathbf{k}\cdot\mathbf{r}) . \quad (17)$$

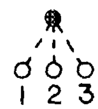
Similarly, we write $m(r_{34})$ as

$$m(r_{34}) = \sum_l M_l(r_{13},r_{14}) P_l(\cos\theta_{314})$$
$$= \sum_{l,m} \frac{4\pi}{2l+1} M_l(r_{13},r_{14}) Y_{lm}^*(\omega_{213}) Y_{lm}(\omega_{214}) , \quad (18)$$

using the addition theorem for spherical harmonics[25] in the second equality to bring out the specific angular variables needed. Employing the orthogonality of the spherical harmonics, we get

$$\int d\omega_{214}\, m(r_{24}) m(r_{34})$$
$$= \sum_l \frac{4\pi}{2l+1} M_l(r_{12},r_{14}) M_l(r_{13},r_{14}) P_l(\cos\theta_{213}) , \quad (19)$$

and hence

$$J\left(\right.$$  $$\left.\right) = \frac{8\pi}{81} \int_0^\infty dr\, r^2 m(r) \left(\int_0^\infty \frac{ds}{s} M_4(s,r) \right)^2 . \quad (20)$$

To calculate the second integral above, we may use (16) and the fact that

$$\tilde{m}(k) = (4\pi a^2/k) j_1(ka) \quad (21)$$

to obtain M_4. Thus, we find

$$\int_0^\infty \frac{ds}{s} M_4(s,r) = \frac{18a^2}{\pi} \int_0^\infty dk\, k j_1(ka) j_4(kr) \int_0^\infty \frac{ds}{s} j_4(ks)$$
$$= \frac{12a^2}{5\pi} \int_0^\infty dk\, k j_1(ka) j_4(kr)$$
$$= \left[3\left(\frac{a}{r}\right)^3 - \frac{21}{5}\left(\frac{a}{r}\right)^5 \right] H(r-a) , \quad (22)$$

where $H(x)$ is the Heaviside unit function

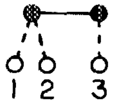
$$H(x) = 0, \quad x<0 ,$$
$$= 1, \quad x>0 , \quad (23)$$

and the integrals involving the j_l's may be found in Ref. 26. Now we can see that the contribution of the diagram in (11a) to J is zero because (20) has conflicting step-function requirements in its two integrals. Thus, we find that

$$J[S_3^{(1)}] = 0 . \quad (24)$$

B. Evaluation of $J[S_3^{(2)}]$

To simplify the contribution of the diagrams in (11b), we utilize the freedom afforded by the homogeneity and isotropy of the system to conveniently choose the origin and orientation of the coordinate frame. If we first choose the origin at $\mathbf{r}_4$, then we find for the first term of $S_3^{(2)}$ in (11b) that

$$J\left(\frac{1}{V_1^2}\right.$$  $$\left.\right) = \frac{1}{8\pi^2 V_1^2}\int d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3 \frac{P_4(\cos\theta_{213})}{r_{12}^3 r_{13}^3}\int d\mathbf{r}_5\, m(r_{14})m(r_{24})g(r_{45})m(r_{35})$$

$$= \frac{1}{8\pi^2}\int d\mathbf{r}_5\, g(r_{45})W_1(r_{45})\,, \quad (25)$$

where

$$W_1(r_{45}) = \frac{1}{V_1^2}\int d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3 \frac{P_4(\cos\theta_{213})}{r_{12}^3 r_{13}^3} m(r_{14})m(r_{24})m(r_{35})\,. \quad (26)$$

Using (18) for the expansion of $m(r_{24})$ and $m(r_{35})$, and then using the completeness relation for the Y_{lm}'s, we get

$$W_1(r_{45}) = \frac{1}{V_1^2}\int d\mathbf{r}_1\, d\mathbf{r}_2\, m(r_{14})m(r_{24})\frac{1}{r_{12}^3}\sum_{l,m}\frac{4\pi}{2l+1}\int_0^\infty \frac{dr_{13}}{r_{13}} M_l(r_{13},r_{15})\int d\omega_{213}\, P_4(\cos\theta_{213})Y^*_{lm}(\omega_{213})Y_{lm}(\omega_{215})$$

$$= \frac{1}{V_1^2}\int d\mathbf{r}_1\, m(r_{14})\left(\frac{4\pi}{9}\int_0^\infty \frac{dx}{x}M_4(x,r_{15})\right)\int d\mathbf{r}_2 \frac{P_4(\cos\theta_{215})}{r_{12}^3} m(r_{24})$$

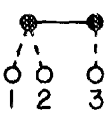
$$= \frac{1}{V_1^2}\int d\mathbf{r}_1\, m(r_{14})P_4(\cos\theta_{415})\left(\frac{4\pi}{9}\int_0^\infty \frac{dx}{x}M_4(x,r_{15})\right)\left(\frac{4\pi}{9}\int_0^\infty \frac{dy}{y}M_4(y,r_{14})\right)$$

$$= \frac{1}{V_1^2}\int d\mathbf{r}_1\, H(a-r_{14})P_4(\cos\theta_{415})\left\{\frac{4}{3}\pi\left[\left(\frac{a}{r_{15}}\right)^3 - \frac{7}{5}\left(\frac{a}{r_{15}}\right)^5\right]H(r_{15}-a)\right\}$$

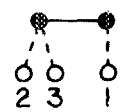
$$\times\left\{\frac{4}{3}\pi\left[\left(\frac{a}{r_{14}}\right)^3 - \frac{7}{5}\left(\frac{a}{r_{14}}\right)^5\right]H(r_{14}-a)\right\}$$

$$= 0\,, \quad (27)$$

because of the conflicting demands of the step functions on r_{14}. In deriving (27), we have made use of (22). Thus

$$J\left(\frac{1}{V_1^2}\right.$$  $$\left.\right) = 0\,. \quad (28)$$

Now interchanging labels 2 and 3 in the procedure above gives the same integral, and hence the contribution of the second diagram of (11b) to $J[S_3^{(2)}]$ is also zero. But such is not the case for the last diagram of (11b). Proceeding as above, one obtains

$$J\left(\frac{1}{V_1^2}\right.$$  $$\left.\right) = \frac{1}{8\pi^2}\int d\mathbf{r}_5\, g(r_{45})W_2(r_{45})\,, \quad (29)$$

where

$$W_2(r_{45}) = \frac{1}{V_1^2}\int d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3 \frac{P_4(\cos\theta_{213})}{r_{12}^3 r_{13}^3} m(r_{15})m(r_{24})m(r_{34})$$

$$= \frac{4\pi}{a^6}\int_0^\infty dr_{14}\, r_{14}^2\, M_0(r_{14},r_{45})\left[\left(\frac{a}{r_{14}}\right)^3 - \frac{7}{5}\left(\frac{a}{r_{14}}\right)^5\right]^2 H(r_{14}-a)\,. \quad (30)$$

In obtaining (30) above we have made repeated use of (22). Next, using the inverse expansion of (15) to write M_0 and making a change of variables back to r_{15}, we get

$$M_0(r_{14},r_{45}) = \frac{1}{2}\int_{-1}^{1} d(\cos\theta)$$

$$\times m[(r_{14}^2 + r_{45}^2 - 2r_{14}r_{45}\cos\theta)^{1/2}]$$

$$= \frac{1}{2}\int_{|r_{14}-r_{45}|}^{r_{14}+r_{45}} dr_{15}\frac{r_{15}}{r_{14}r_{45}} m(r_{15})\,. \quad (31)$$

Use of (31) in (30) leads to

$$W_2(t) = A - B + C\,, \quad (32)$$

where

$$\begin{Bmatrix} A \\ B \\ C \end{Bmatrix} = \begin{Bmatrix} 1 \\ \frac{14}{5}a^2 \\ \frac{49}{25}a^4 \end{Bmatrix}\frac{2\pi}{t}\int_a^\infty dr \begin{Bmatrix} \frac{1}{r^5} \\ \frac{1}{r^7} \\ \frac{1}{r^9} \end{Bmatrix}\int_{|r-t|}^{r+t} ds\, sm(s)\,. \quad (33)$$

Now, if we perform the two integrals in (33), we finally obtain

$$J[S_3^{(2)}] = \frac{1}{2\pi}\int_\sigma^\infty dr\, r^2 g(r)W_2(r)\,, \quad (34)$$

where for $r \geqslant \sigma$

$$W_2(r) = \frac{4}{3}\pi\left(\frac{a^3}{(r^2-a^2)^3} - \frac{14}{5}\frac{a^5}{(r^2-a^2)^4} - \frac{63}{25}\frac{a^7}{(r^2-a^2)^5} + \frac{196}{25}\frac{a^9}{(r^2-a^2)^6} + \frac{168}{25}\frac{a^{11}}{(r^2-a^2)^7}\right). \quad (35)$$

Here $\sigma = 2a$ is the sphere diameter and we have used the fact that $g(r) = 0$ for $r < \sigma$. It may be noted that in the low-density limit $\rho \to 0$, $g(r) \to 1$ for $r > \sigma$, and then (34) reduces to

$$\lim_{\rho\to 0} J[S_3^{(2)}] = \tfrac{7213}{109\,350} - \tfrac{1}{24}\ln 3 . \quad (36)$$

The importance of this result lies in the fact that if we write a low-density expansion of the parameter η_2 as

$$\eta_2 = f_1\phi_2 + O(\phi_2^2) ; \quad (37)$$

one can then show that

$$f_1 = \lim_{\rho\to 0} \tfrac{45}{42} I[S_3^{(2)}] + \lim_{\rho\to 0} \tfrac{150}{7} J[S_3^{(2)}] . \quad (38)$$

In a previous paper[27] it has been shown that

$$\lim_{\rho\to 0} I[S_3^{(2)}] = \tfrac{5}{54} - \tfrac{1}{24}\ln 3 . \quad (39)$$

Combining (36) and (39) in (38), one finds that $f_1 = 0.482\,74$, which is the result quoted in the previously mentioned paper[19] on the third-order bounds on shear modulus in the dilute limit.

C. Evaluation of $J[S_3^{(3)}]$

To simplify the contribution of the diagram in (11c), we employ the same technique used in the previous subsection. Thus, choosing the origin at $\mathbf{r}_4$, we find that

$$J[S_3^{(3)}] = \frac{1}{16\pi^2}\int d\mathbf{r}_5\, d\mathbf{r}_6\, g_3(r_{45},r_{46},r_{56}) \times Q(r_{45},r_{46},r_{56}) , \quad (40)$$

where

$$Q(r_{45},r_{46},r_{56}) = \frac{2}{V_1^3}\int d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3\, m(r_{14})m(r_{25}) \times m(r_{36})\frac{P_4(\cos\theta_{213})}{r_{12}^3 r_{13}^3} . \quad (41)$$

The integrals over $\mathbf{r}_3$ first and then $\mathbf{r}_2$ are done by following the same method as applied to simplify (26) to the form (27). The result in this case is

$$\int d\mathbf{r}_2\, m(r_{25})\int d\mathbf{r}_3\, m(r_{36})\frac{P_4(\cos\theta_{213})}{r_{12}^3\, r_{13}^3} = V_1^2\, H(r_{15}-a)H(r_{16}-a)\left[1-\frac{7}{5}\left(\frac{a}{r_{15}}\right)^2\right]\left[1-\frac{7}{5}\left(\frac{a}{r_{16}}\right)^2\right]\frac{P_4(\cos\theta_{516})}{r_{15}^3 r_{16}^3} . \quad (42)$$

In Eqs. (40) and (41), $r_{14} < a$ and $r_{46} > 2a$ because of the m and g functions involved. Thus $H(r_{16}-a)$ is redundant in (42). Similarly, because $r_{14} < a$ and $r_{45} > 2a$, the use of $H(r_{15}-a)$ is redundant as well. Thus, we drop these H functions, and Eq. (41) is simplified to

$$Q(r_{45},r_{46},r_{56}) = \frac{2}{V_1}\int d\mathbf{r}_1\, m(r_{14})\left[1-\frac{7}{5}\left(\frac{a}{r_{15}}\right)^2\right] \times\left[1-\frac{7}{5}\left(\frac{a}{r_{16}}\right)^2\right]\frac{P_4(\cos\theta_{516})}{r_{15}^3 r_{16}^3} . \quad (43)$$

The difficulty in simplifying this expression any further lies in explicitly bringing out the orientation dependence of the integrand for the final integration over $\mathbf{r}_1$. For this we shall use the coordinate frame arrangement shown in Fig. 1. With respect to this figure, we use the identity

$$\cos\theta_{516} = \cos\theta_{415}\cos\theta_{416} + \sin\theta_{415}\sin\theta_{416}\cos\psi , \quad (44)$$

where ψ is the angle between the planes 541 and 641, to write the addition theorem expansion for $P_4(\cos\theta_{516})$ as

$$P_4(\cos\theta_{516}) = \sum_{m=0}^{4}\alpha_m\frac{(4-m)!}{(4+m)!}P_4^m(\cos\theta_{415}) \times P_4^m(\cos\theta_{416})\cos(m\psi) , \quad (45)$$

where

$$\alpha_m = 1, \quad m = 0 ,$$
$$= 2, \quad m > 0 . \quad (46)$$

Using (45) in (43), we find that

$$Q(r_{45},r_{46},r_{56}) = \frac{2}{V_1}\sum_{m=0}^{4}\alpha_m\frac{(4-m)!}{(4+m)!}\int_0^a dr_{14}\, r_{14}^2\int d\omega_{641}\left\{\frac{P_4^m(\cos\theta_{415})}{r_{15}^3}\left[1-\frac{7}{5}\left(\frac{a}{r_{15}}\right)^2\right]\right\} \times\left\{\frac{P_4^m(\cos\theta_{416})}{r_{16}^3}\left[1-\frac{7}{5}\left(\frac{a}{r_{16}}\right)^2\right]\right\}\cos(m\psi) . \quad (47)$$

We show in Appendix B that each of the expressions within large brackets in (47) can be expanded in terms of the corresponding opposite angles at the base of the coordinate frame, giving

$$Q(r_{45},r_{46},r_{56}) = \frac{2}{V_1}\sum_{m=0}^{4}\alpha_m\frac{(4-m)!}{(4+m)!}\int_0^a dr_{14}\, r_{14}^2\int d\omega_{641} \times\sum_{l,l'}\left(\frac{r_{14}^{l-4}}{r_{45}^{l-1}}\beta_{lm} - \frac{r_{14}^{l-2}}{r_{45}^{l+1}}\gamma_{lm} - \frac{7}{5}a^2\frac{r_{14}^{l-4}}{r_{45}^{l+1}}\nu_{lm}\right)P_l^m(\cos\theta_{541}) \times\left(\frac{r_{14}^{l'-4}}{r_{46}^{l'-1}}\beta_{l'm} - \frac{r_{14}^{l'-2}}{r_{46}^{l'+1}}\gamma_{l'm} - \frac{7}{5}a^2\frac{r_{14}^{l'-4}}{r_{46}^{l'+1}}\nu_{l'm}\right)P_{l'}^m(\cos\theta_{641})\cos(m\psi) , \quad (48)$$

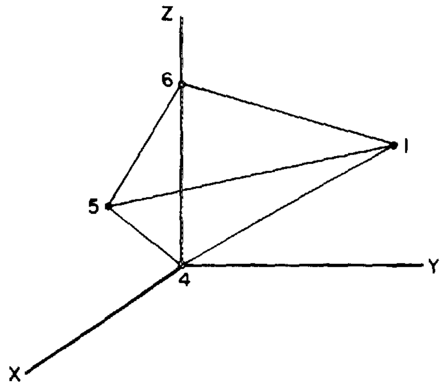


FIG. 1. Coordinate system for Eq. (43).

where

$$\beta_{l0} = \frac{7(l-3)(l-2)(l-1)l}{24(2l-1)},$$
$$\beta_{l1} = -\frac{7(l-3)(l-2)(l-1)}{6(2l-1)},$$
$$\beta_{l2} = \frac{7(l-3)(l-2)}{2(2l-1)}, \quad (49)$$
$$\beta_{l3} = -\frac{7(l-3)}{2l-1},$$
$$\beta_{l4} = \frac{7}{2l-1},$$

$$\gamma_{l0} = \frac{(l-2)(l-1)l(7l+11)}{24(2l+3)},$$
$$\gamma_{l1} = -\frac{(l-2)(l-1)(7l+9)}{6(2l+3)},$$
$$\gamma_{l2} = \frac{(l-2)(7l+3)}{2(2l+3)}, \quad (50)$$
$$\gamma_{l3} = -\frac{7(l-1)}{2l+3},$$
$$\gamma_{l4} = \frac{7}{2l+3},$$

and

$$\nu_{lm} = \left(\frac{2l-1}{7}\right)\beta_{lm}. \quad (51)$$

To be able to do the angular integral in (48), we rotate the coordinate frame of Fig. 1 by an angle θ_{641} about an axis perpendicular to the $(\hat{\mathbf{r}}_{46}, \hat{\mathbf{r}}_{41})$ plane with a temporary reassignment of the coordinate frame so that $\hat{\mathbf{r}}_{41}$ is in the (x,z) plane as in Fig. 2. The unit vector $\hat{\mathbf{r}}_{45}$ has an orientation (θ_{546},ϕ) with respect to the (x,y,z) frame and (θ_{541},ψ) with respect to the rotated (x',y',z') frame, because ψ is the angle between the (x,z) plane and the $(\hat{\mathbf{r}}_{41},\hat{\mathbf{r}}_{51})$ plane. The Euler angles of rotation between the frames are $(0,\theta_{641},0)$. Thus the transformation theorem for this special case is[25]

$$Y_{lm}(\theta_{541},\psi) = \sum_{m'} d^{\,l}_{m'm}(\theta_{641})\, Y_{lm'}(\theta_{546},\phi). \quad (52)$$

Writing out the spherical harmonics on both the sides in terms of the associated Legendre functions, we get

$$P_l^m(\cos\theta_{541})\cos(m\psi) = \left(\frac{(l+m)!}{(l-m)!}\right)^{1/2} \sum_{m'=0}^{l} \alpha_{m'}\,(-1)^{m'-m}\left(\frac{(l-m')!}{(l+m')!}\right)^{1/2} d^{\,l}_{m'm}(\theta_{641}) P_l^{m'}(\cos\theta_{546})\cos(m'\phi). \quad (53)$$

Using this, we find the angular integral in (48) to be

$$\int d(\cos\theta_{641})\, d\phi \left[P_{l'}^m(\cos\theta_{541})\cos(m\psi)\right] P_l^m(\cos\theta_{641}) = \frac{4\pi}{2l+1}\,\frac{(l+m)!}{(l-m)!}\, P_l(\cos\theta_{546})\delta_{l'l}, \quad (54)$$

and thus (48) reduces to

$$Q(r_{45},r_{46},r_{56}) = \frac{2}{V_1}\sum_{m=0}^{4}\alpha_m \frac{(4-m)!}{(4+m)!}\int_0^a dr_{14}\, r_{14}^2 \sum_l \frac{4\pi}{2l+1}\,\frac{(l+m)!}{(l-m)!}\left(\frac{r_{14}^{l-4}}{r_{45}^{l-1}}\beta_{lm} - \frac{r_{14}^{l-2}}{r_{45}^{l+1}}\gamma_{lm} - \frac{7}{5}a^2\frac{r_{14}^{l-4}}{r_{45}^{l+1}}\nu_{lm}\right)$$
$$\times\left(\frac{r_{14}^{l-4}}{r_{46}^{l-1}}\beta_{lm} - \frac{r_{14}^{l-2}}{r_{46}^{l+1}}\gamma_{lm} - \frac{7}{5}a^2\frac{r_{14}^{l-4}}{r_{46}^{l+1}}\nu_{lm}\right) P_l(\cos\theta_{546}). \quad (55)$$

Next we compute the integral over r_{14} to obtain

$$Q(r_{45},r_{46},r_{56}) = \frac{2}{V_1}\sum_{m=0}^{4}\alpha_m \frac{(4-m)!}{(4+m)!}\sum_l \frac{4\pi}{2l+1}\,\frac{(l+m)!}{(l-m)!}\left[\frac{a^{2l-1}}{r_{45}^{l+1}r_{46}^{l+1}}\left(\frac{\gamma_{lm}^2}{2l-1} + \frac{14}{5}\,\frac{\gamma_{lm}\nu_{lm}}{2l-3} + \frac{49}{25}\,\frac{\nu_{lm}^2}{2l-5}\right)\right.$$
$$\left. - a^{2l-3}\left(\frac{1}{r_{45}^{l+1}r_{46}^{l-1}} + \frac{1}{r_{45}^{l-1}r_{46}^{l+1}}\right)\left(\frac{\beta_{lm}\gamma_{lm}}{2l-3} + \frac{7}{5}\,\frac{\beta_{lm}\nu_{lm}}{2l-5}\right) + \frac{a^{2l-5}}{r_{45}^{l-1}r_{46}^{l-1}}\,\frac{\beta_{lm}^2}{2l-5}\right] P_l(\cos\theta_{546}). \quad (56)$$

In the above we complete the sum over m, using (46) and (49)–(51), and then after a considerable amount of algebra, find that

$$Q(r_{45},r_{46},r_{56}) = \frac{14}{5!}\sum_{l=4}^{\infty} l(l-1)(l-2)(l-3)\,\frac{(2l-3)}{(2l-1)}\,\frac{a^{2l-8}}{r_{45}^{l-1}r_{46}^{l-1}}$$
$$\times\left[1 - \frac{2}{5}(l+1)\left(\frac{2l-1}{2l-3}\right)\left(\frac{a}{r_{45}}\right)^2\right]\left[1 - \frac{2}{5}(l+1)\left(\frac{2l-1}{2l-3}\right)\left(\frac{a}{r_{46}}\right)^2\right] P_l(\cos\theta_{546})$$
$$+ \frac{8}{5!}\sum_{l=3}^{\infty}\frac{l(l-1)(l-2)(11l+15)}{(2l+3)(2l-3)}\,\frac{a^{2l-4}}{r_{45}^{l+1}r_{46}^{l+1}} P_l(\cos\theta_{546}). \quad (57)$$

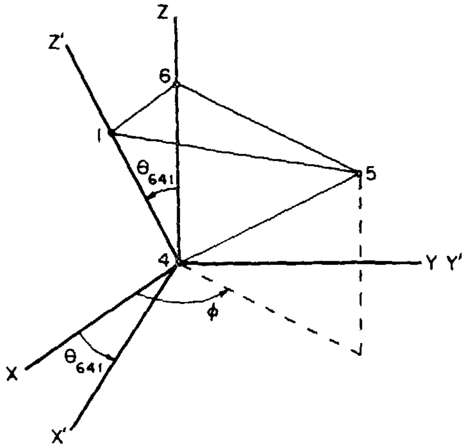


FIG. 2. Coordinate system for Eq. (52).

The result (57), when substituted in (40), provides the final simplified form of $J[S_3^{(3)}]$. Since the only orientation dependence in $Q(r_{45},r_{46},r_{56})$ comes through $P_l(\cos\theta_{546})$, that is, through the angle between $\mathbf{r}_{45}$ and $\mathbf{r}_{46}$, it is clear that if we replace g_3 in (40) by $g(r_{45})g(r_{46})$, then the integral would vanish identically. But, whereas the integral (40) is conditionally convergent, depending upon the coordinate system and the order of performing the integration, the subtraction of the $g(r_{45})g(r_{46})$ term from g_3 before doing the integral makes it absolutely convergent. This fact was already remarked upon following Eq. (9).

If we combine the results from the previous three subsections, namely, (24) and (34) along with (35), and (40) along with (57), then the key integral $J[\hat{S}_3]$ takes, the form

$$J[\hat{S}_3] = \frac{1}{8\pi^2}\phi_2^2 \int d\mathbf{r}\, g(r) W_2(r) + \frac{\phi_2^3}{16\pi^2}\int d\mathbf{r}_2\, d\mathbf{r}_3 [g_3(r_{12},r_{13},r_{23}) - g(r_{12})g(r_{13})] Q(r_{12},r_{13},r_{23}) , \quad (58)$$

where the function Q is given in (57) and where $W_2(r)$ is given in (35). Finally, for completeness, we end this section by giving the simplified form of the other key integral $I[\hat{S}_3]$ as obtained by Lado and Torquato[20]:

$$I[\hat{S}_3] = \frac{2}{3}\phi_2^2 a^3 \int_{2a}^{\infty} dr \frac{r^2 g(r)}{(r^2-a^2)^3} + \frac{\phi_2^3}{16\pi^2}\sum_{l=2}^{\infty} l(l-1)a^{2l-4}\int d\mathbf{r}_2\, d\mathbf{r}_3 [g_3(r_{12},r_{13},r_{23}) - g(r_{12})g(r_{13})] \frac{P_l(\cos\theta_{213})}{r_{12}^{l+1} r_{13}^{l+1}} . \quad (59)$$

IV. CONCLUSIONS

For the model of impenetrable equisized spherical inclusions randomly distributed throughout a matrix, we have now simplified expressions for the two key integrals $I[\hat{S}_3]$ and $J[\hat{S}_3]$ that arise in the third-order McCoy and MPT bounds on the effective shear modulus G_e. It may again be noted that the simplification of $I[\hat{S}_3]$ was done in a previous paper.[20] Both of these tasks were accomplished by expanding orientation-dependent terms in the two integrands in spherical harmonics and utilizing the orthogonality properties of this basis set. The resulting simplified integrals are shown to depend upon the one-, two-, and three-body distribution functions. We believe that this is the first time that the key integral $J[\hat{S}_3]$, required for the third-order bounds on G_e, has been simplified to this extent. In a subsequent paper we shall employ this simplified form for J to compute the McCoy and MPT bounds for a wide range of sphere volume fractions.

ACKNOWLEDGMENTS

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APPENDIX A

As already used in (15), and following Barker and Monaghan,[24] we expand angle-dependent functions $f(r_{23})$, which are well behaved (i.e., functions with a finite number of finite discontinuities), in Legendre polynomials:

$$f(r_{23}) = \sum_{l=0}^{\infty} F_l(r_{12},r_{13}) P_l \cos(\theta_{213}) , \quad (A1)$$

where the orthogonality of the Legendre polynomials lead to the inverse expansion

$$F_l(r_{12},r_{13}) = \frac{2l+1}{2}\int_{-1}^{1} d(\cos\theta_{213}) f(r_{23}) P_l(\cos\theta_{213}) , \quad (A2)$$

and

$$r_{23}^2 = r_{12}^2 + r_{13}^2 - 2r_{12}r_{13}\cos\theta_{213} . \quad (A3)$$

But $f(r_{23})$ may also be expanded in plane waves as

$$f(r_{23}) = \frac{1}{(2\pi)^3}\int d\mathbf{k}\, \tilde{f}(k) \exp(i\mathbf{k}\cdot\mathbf{r}_{23}) , \quad (A4)$$

where the Fourier transform $\tilde{f}(k)$ is given by an expression similar to (17). If we now arrange that $\mathbf{r}_1$ is the origin, $\hat{\mathbf{r}}_{12}$ is along the z axis, the $(\hat{\mathbf{r}}_{12},\hat{\mathbf{r}}_{13})$ plane is the (x,z) plane, and let (θ,ϕ) be the angular coordinates of the wave vector $\mathbf{k}$ in this frame, then the well-known expansion of plane waves in spherical waves gives[28]

$$\exp(i\mathbf{k}\cdot\mathbf{r}_{23}) = \exp(i\mathbf{k}\cdot\mathbf{r}_{13})\exp(-ikr_{12}\cos\theta) = \left(4\pi\sum_{l,m} i^l j_l(kr_{13}) Y_{lm}^{*}(\theta_{213},0) Y_{lm}(\theta,\phi)\right)\left(\sum_{l'}(2l'+1)(-i)^{l'} j_{l'}(kr_{12}) P_{l'}(\cos\theta)\right) . \quad (A5)$$

Then, using (A5) in the plane-wave expansion of $f(r_{23})$, one gets

$$f(r_{23}) = \frac{1}{2\pi^2} \sum_{l=0}^{\infty} (2l+1) P_l(\cos\theta_{213}) \times \int_0^\infty dk\, k^2 \tilde{f}(k) j_l(kr_{12}) j_l(kr_{13}) . \quad (A6)$$

Comparison of (A6) with (A1) gives

$$F_l(r_{12}, r_{13}) = \frac{2l+1}{2\pi^2} \int_0^\infty dk\, k^2 \tilde{f}(k) j_l(kr_{12}) j_l(kr_{13}) , \quad (A7)$$

a result we made use of in (20).

APPENDIX B

This appendix deals with the specific task of rewriting $P_4^m(\cos\phi)/t^3$ and $P_4^m(\cos\phi)/t^5$ (see Fig. 3) in terms of r, s, and $\cos\theta$ for $m = 0,1,2,3$, and 4, and the condition $s < r$. We start with the generating function of the Legendre polynomials,[29]

$$\frac{r}{t} = \left(1 - 2\frac{s}{r}\cos\theta + \frac{s^2}{r^2}\right)^{-1/2} = \sum_{l=0}^{\infty} \left(\frac{s}{r}\right)^l P_l(\cos\theta) . \quad (B1a)$$

Successive differentiation of (B1a) with respect to $\cos\theta$, denoted by primes, gives

$$\left(\frac{r}{t}\right)^3 = \sum_l \left(\frac{s}{r}\right)^{l-1} P_l'(\cos\theta) , \quad (B1b)$$

$$\left(\frac{r}{t}\right)^5 = \frac{1}{3} \sum_l \left(\frac{s}{r}\right)^{l-2} P_l''(\cos\theta) , \quad (B1c)$$

$$\left(\frac{r}{t}\right)^7 = \frac{1}{15} \sum_l \left(\frac{s}{r}\right)^{l-3} P_l'''(\cos\theta) , \quad (B1d)$$

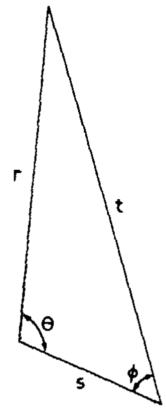

FIG. 3. General geometry considered in Appendix B for transforming the arguments of the Legendre polynomials from $\cos\phi$ to $\cos\theta$.

$$\left(\frac{r}{t}\right)^9 = \frac{1}{105} \sum_l \left(\frac{s}{r}\right)^{l-4} P_l''''(\cos\theta) . \quad (B1e)$$

Next the law of sines gives

$$\sin\phi = (r/t)\sin\theta , \quad (B2a)$$

and this along with $t^2 = r^2 + s^2 - 2rs\cos\theta$ gives

$$\cos\phi = (r/t)(s/r - \cos\theta) . \quad (B2b)$$

For brevity, from now on we will write $x = \cos\theta$ and drop the argument x on P_l and its derivatives; their presence will be assumed implicitly unless otherwise stated [as in $P_l^m(\cos\theta)$]. Also, we will freely use Legendre's equation (along with its two higher-order derivatives) and recurrence relations[30] for simplification. Similar relations for the associated Legendre functions,

$$P_l^m \equiv (1-x^2)^{m/2} d^m P_l(x)/dx^m , \quad (B3)$$

will also be used whenever necessary.

(i) $m = 0$ case

For the $m = 0$ case we have

$$\begin{aligned}
\left(\frac{r}{t}\right)^3 P_4(\cos\phi) &= \frac{35}{8}\left(\frac{r}{t}\right)^7 \sin^4\theta - 5\left(\frac{r}{t}\right)^5 \sin^2\theta + \left(\frac{r}{t}\right)^3 \\
&= \sum_l \left(\frac{s}{r}\right)^{l-3} \left(\frac{7}{24}(1-x^2)^2 P_l''' - \frac{5}{3}(1-x^2) P_{l-1}'' + P_{l-2}'\right) \\
&= \sum_l \left(\frac{s}{r}\right)^{l-3} \frac{1}{24} \frac{(l-1)(l-2)}{(2l+1)} [7l(l+1)P_{l+1} - (l-3)(7l+4)P_{l-1}] \\
&= \sum_l \left[\left(\frac{s}{r}\right)^{l-4} \frac{7l(l-1)(l-2)(l-3)}{24(2l-1)} - \left(\frac{s}{r}\right)^{l-2} \frac{l(l-1)(l-2)(7l+11)}{24(2l+3)}\right] P_l \quad (B4)
\end{aligned}$$

and

$$\begin{aligned}
\left(\frac{r}{t}\right)^5 P_4(\cos\phi) &= \frac{35}{8}\left(\frac{r}{t}\right)^9 \sin^4\theta - 5\left(\frac{r}{t}\right)^7 \sin^2\theta + \left(\frac{r}{t}\right)^5 \\
&= \sum_l \left(\frac{s}{r}\right)^{l-3} \left(\frac{1}{24}(1-x^2)^2 P_{l+1}'''' - \frac{1}{3}(1-x^2) P_l''' + \frac{1}{3} P_l''\right) \\
&= \sum_l \left(\frac{s}{r}\right)^{l-4} \frac{1}{24} l(l-1)(l-2)(l-3) P_l . \quad (B5)
\end{aligned}$$

Thus we have from (B4) and (B5)

$$\frac{P_4(\cos\phi)}{t^3} = \sum_l \left(\frac{s^{l-4}}{r^{l-1}} \frac{7l(l-1)(l-2)(l-3)}{24(2l-1)} - \frac{s^{l-2}}{r^{l+1}} \frac{l(l-1)(l-2)(7l+11)}{24(2l+3)}\right) P_l(\cos\theta) , \quad (B6)$$

and

$$\frac{P_4(\cos\phi)}{t^5}=\sum_l \frac{s^{l-4}}{r^{l+1}}\frac{1}{24}l(l-1)(l-2)(l-3)P_l(\cos\theta)\,. \tag{B7}$$

(ii) $m=1$ case

For the $m=1$ case we have

$$\left(\frac{r}{t}\right)^3 P_4^1(\cos\phi)=\frac{5}{2}\sin\theta\left(\frac{s}{r}-\cos\theta\right)\left[4\left(\frac{r}{t}\right)^5-7\left(\frac{r}{t}\right)^7\sin^2\theta\right]$$

$$=\frac{5}{2}(1-x^2)^{1/2}\sum_l\left(\frac{s}{r}\right)^{l-3}\left(\frac{4}{3}P''_{l-2}-\frac{4}{3}xP''_{l-1}-\frac{7}{15}(1-x^2)P'''_{l-1}+\frac{7}{15}x(1-x^2)P'''_l\right)$$

$$=\sum_l\left(\frac{s}{r}\right)^{l-3}\frac{(l-2)}{6(2l+1)}\left[(7l+2)(l-3)P^1_{l-1}-7l(l-1)P^1_{l+1}\right]$$

$$=\sum_l\left[\left(\frac{s}{r}\right)^{l-4}\frac{(-7)(l-1)(l-2)(l-3)}{6(2l-1)}-\left(\frac{s}{r}\right)^{l-2}\frac{(-1)(l-1)(l-2)(7l+9)}{6(2l+3)}\right]P_l^1 \tag{B8}$$

and

$$\left(\frac{r}{t}\right)^5 P_4^1(\cos\phi)=\frac{5}{2}\sin\theta\left(\frac{s}{r}-\cos\theta\right)\left[4\left(\frac{r}{t}\right)^7-7\left(\frac{r}{t}\right)^9\sin^2\theta\right]$$

$$=(1-x^2)^{1/2}\sum_l\left(\frac{s}{r}\right)^{l-3}\left(\frac{2}{3}xP'''_{l-1}-\frac{2}{3}xP'''_l-\frac{1}{6}(1-x^2)P''''_l+\frac{1}{6}x(1-x^2)P''''_{l+1}\right)$$

$$=\sum_l\left(\frac{s}{r}\right)^{l-4}\left(-\frac{1}{6}\right)(l-1)(l-2)(l-3)P_l^1\,. \tag{B9}$$

Thus (B8) and (B9) give the desired relations

$$\frac{P_4^1(\cos\phi)}{t^3}=-\sum_l\left(\frac{s^{l-4}}{r^{l-1}}\frac{7(l-1)(l-2)(l-3)}{6(2l-1)}-\frac{s^{l-2}}{r^{l+1}}\frac{(l-1)(l-2)(7l+9)}{6(2l+3)}\right)P_l^1(\cos\theta) \tag{B10}$$

and

$$\frac{P_4^1(\cos\phi)}{t^5}=-\sum_l\frac{s^{l-4}}{r^{l+1}}\frac{1}{6}(l-1)(l-2)(l-3)P_l^1(\cos\theta)\,. \tag{B11}$$

(iii) $m=2$ case

For the $m=2$ case we have

$$\left(\frac{r}{t}\right)^3 P_4^2(\cos\phi)=\frac{15}{2}\sin^2\theta\left[6\left(\frac{r}{t}\right)^5-7\left(\frac{r}{t}\right)^7\sin^2\theta\right]$$

$$=\frac{15}{2}(1-x^2)\sum_l\left(\frac{s}{r}\right)^{l-3}\left(2P''_{l-1}-\frac{7}{15}(1-x^2)P'''_l\right)$$

$$=\sum_l\left(\frac{s}{r}\right)^{l-3}\frac{1}{2(2l+1)}\left(7(l-1)(l-2)P^2_{l+1}-(7l-4)(l-3)P^2_{l-1}\right)$$

$$=\sum_l\left[\left(\frac{s}{r}\right)^{l-4}\frac{7(l-2)(l-3)}{2(2l-1)}-\left(\frac{s}{r}\right)^{l-2}\frac{(l-2)(7l+3)}{2(2l+3)}\right]P_l^2 \tag{B12}$$

and

$$\left(\frac{r}{t}\right)^5 P_4^2(\cos\phi)=\frac{15}{2}\sin^2\theta\left[6\left(\frac{r}{t}\right)^7-7\left(\frac{r}{t}\right)^9\sin^2\theta\right]$$

$$=(1-x^2)\sum_l\left(\frac{s}{r}\right)^{l-3}\left(3P'''_l-\frac{1}{2}(1-x^2)P''''_{l+1}\right)$$

$$=\sum_l\left(\frac{s}{r}\right)^{l-4}\frac{1}{2}(l-2)(l-3)P_l^2\,. \tag{B13}$$

Thus

$$\frac{P_4^2(\cos\phi)}{t^3}=\sum_l\left(\frac{s^{l-4}}{r^{l-1}}\frac{7(l-2)(l-3)}{2(2l-1)}-\frac{s^{l-2}}{r^{l+1}}\frac{(l-2)(7l+3)}{2(2l+3)}\right)P_l^2(\cos\theta)\,, \tag{B14}$$

and

$$\frac{P_4^2(\cos\phi)}{t^5}=\sum_l \frac{s^{l-4}}{r^{l+1}}\frac{1}{2}(l-2)(l-3)P_l^2(\cos\theta). \tag{B15}$$

(iv) $m=3$ case

For the $m=3$ case we have

$$\begin{aligned}\left(\frac{r}{t}\right)^3 P_4^3(\cos\phi) &= 105\sin^3\theta\left(\frac{s}{r}-\cos\theta\right)\left(\frac{r}{t}\right)^7\\ &= 7(1-x^2)^{3/2}\sum_l\left(\frac{s}{r}\right)^{l-3}(P'''_{l-1}-xP'''_l)\\ &= \sum\left(\frac{s}{r}\right)^{l-3}\frac{7(l-2)}{2l+1}(P^3_{l-1}-P^3_{l+1})\\ &= -\sum_l\left[\left(\frac{s}{r}\right)^{l-4}\frac{7(l-3)}{2l-1}-\left(\frac{s}{r}\right)^{l-2}\frac{7(l-1)}{2l+3}\right]P_l^3\end{aligned} \tag{B16}$$

and

$$\begin{aligned}\left(\frac{r}{t}\right)^5 P_4^3(\cos\phi) &= 105\sin^3\theta\left(\frac{s}{r}-\cos\theta\right)\left(\frac{r}{t}\right)^9\\ &= (1-x^2)^{3/2}\sum_l\left(\frac{s}{r}\right)^{l-3}(P''''_l-xP''''_{l+1})\\ &= -\sum_l\left(\frac{s}{r}\right)^{l-4}(l-3)P_l^3.\end{aligned} \tag{B17}$$

Thus

$$\frac{P_4^3(\cos\phi)}{t^3}=-\sum_l\left(\frac{s^{l-4}}{r^{l-1}}\frac{7(l-3)}{2l-1}-\frac{s^{l-2}}{r^{l+1}}\frac{7(l-1)}{2l+3}\right)P_l^3(\cos\theta) \tag{B18}$$

and

$$\frac{P_4^3(\cos\phi)}{t^5}=-\sum_l\frac{s^{l-4}}{r^{l+1}}(l-3)P_l^3(\cos\theta). \tag{B19}$$

(v) $m=4$ case

For the $m=4$ case we have

$$\begin{aligned}\left(\frac{r}{t}\right)^3 P_4^4(\cos\phi) &= 105\left(\frac{r}{t}\right)^7\sin^4\theta\\ &= 7(1-x^2)^2\sum_l\left(\frac{s}{r}\right)^{l-3}P'''_l\\ &= \sum_l\left(\frac{s}{r}\right)^{l-3}\frac{7}{2l+1}(P^4_{l+1}-P^4_{l-1})\\ &= \sum_l\left[\left(\frac{s}{r}\right)^{l-4}\frac{7}{(2l-1)}-\left(\frac{s}{r}\right)^{l-2}\frac{7}{2l+3}\right]P_l^4\end{aligned} \tag{B20}$$

and

$$\begin{aligned}\left(\frac{r}{t}\right)^5 P_4^4(\cos\phi) &= 105\left(\frac{r}{t}\right)^9\sin^4\theta\\ &= (1-x^2)^2\sum_l\left(\frac{s}{r}\right)^{l-4}P''''_l\\ &= \sum_l\left(\frac{s}{r}\right)^{l-4}P_l^4.\end{aligned} \tag{B21}$$

Thus

$$\frac{P_4^4(\cos\phi)}{t^3}=\sum_l\left(\frac{s^{l-4}}{r^{l-1}}\frac{7}{2l-1}-\frac{s^{l-2}}{r^{l+1}}\frac{7}{2l+3}\right)P_l^4(\cos\theta) \tag{B22}$$

and

$$\frac{P_4^4(\cos\phi)}{t^5}=\sum_l\frac{s^{l-4}}{r^{l+1}}P_l^4(\cos\theta). \tag{B23}$$

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