Supplementary Material for “Fundamental Limits to Extinction by Metallic Nanoparticles”

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PROOF OF OPTIMAL EIGENVALUE DISTRIBUTION

We will discuss the optimal distribution of eigenvalues. We emphasize that we are only discussing “bright” modes, i.e. eigenvalues with a non-zero dipole moment. They are the only ones that contribute to extinction, and are the only ones subject to the sum rules. We note again that the depolarization factor \( \lambda_n \) are the only ones subject to the sum rules. We note again that the depolarization factor \( L_n \) is related to the eigenvalue \( \lambda_n \) by \( L_n = 1/2 - \lambda_n \), so that in many instances the words can be used interchangeably.

First, we will show that the optimal distribution takes fewer than three distinct eigenvalues. Intuitively, this can be thought of arising from the fact that the sum in Eq. (5) is incoherent. There is no mixing of modal contributions, and aside from meeting the sum rule requirements, having more than one eigenvalue on the “same side” of 1/3 cannot help. One of the eigenvalues will be strictly better, or worse, than the other.

Let us assume that there are at least three distinct eigenvalues, with increasing depolarization factors \( L_1 < L_2 < L_3 \), and non-zero dipole moments \( p_1 \), \( p_2 \), and \( p_3 \). We will show that the ideal distribution always yields at least one of the dipole moments to be zero. The three eigenvalues need only be a subset of all of the eigenvalues (we will hold the other \( p_i \) and \( L_i \) fixed), such that the three satisfy modified sum rules

\[
\begin{align*}
   p_1 + p_2 + p_3 &= c_1 \quad (S.1a) \\
   p_1L_1 + p_2L_2 + p_3L_3 &= c_2 \quad (S.1b)
\end{align*}
\]

where \( c_1 \leq 3 \) and \( c_2 \leq 1 \).

Assume first that both \( L_1, L_2 \leq 1/3 \). Then we can set \( p_1 \) as an independent variable \( x \) (i.e. \( p_1 = x \)) in the interval \([0, \frac{c_1L_3-c_2}{L_3-L_1}]\); varying \( x \) varies \( p_2 \) and \( p_3 \) accordingly:

\[
\begin{align*}
   p_2 &= p_{2,\text{max}} - x\frac{L_3-L_1}{L_3-L_2} \quad (S.2a) \\
   p_3 &= p_{3,\text{min}} + x\frac{L_2-L_1}{L_3-L_2} \quad (S.2b)
\end{align*}
\]

with \( p_{2,\text{max}} = (c_1L_3-c_2)/(L_3-L_2) \) and \( p_{3,\text{min}} = (c_2-c_1L_3)/(L_3-L_2) \). If we define \( f(L_i) \) to be the contribution of a single dipole moment to the extinction per unit volume

\[
   f(L_i) = 3\left(\frac{1}{L_i - \xi}\right) = \frac{\xi_i}{(L_i - \xi_r)^2 + \xi_i^2} \quad (S.3)
\]

then the total extinction from the three eigenvalues is

\[
\begin{align*}
   \frac{\sigma_{\text{ext}}}{V} &= x f(L_1) + \left(p_{2,\text{max}} - x\frac{\Delta_{31}}{\Delta_{32}}\right) f(L_2) \\
   &+ \left(p_{3,\text{min}} + x\frac{\Delta_{21}}{\Delta_{32}}\right) f(L_3) \quad (S.4)
\end{align*}
\]

where \( \Delta_{ij} = L_i - L_j \). The derivative of the extinction with respect to \( x \),

\[
\frac{\partial}{\partial x} \left(\frac{\sigma_{\text{ext}}}{V}\right) = f(L_1) + \frac{\Delta_{21}f(L_3) - \Delta_{31}f(L_2)}{\Delta_{32}} \quad (S.5)
\]

is a constant with respect to \( x \), meaning the optimal \( x \) is obtained on the boundary: either \( x = 0 \) (\( p_1 = 0 \)), or \( x = x_{\text{max}} \), in which case \( p_2 = 0 \). Depending on the sign of \( \Delta_{21}f(L_3) - \Delta_{31}f(L_2) \), either \( L_1 \) or \( L_2 \) could be optimal; mixing dipole moments among both cannot be.

The math works out identically in the case that both \( L_2 \) and \( L_3 \geq 1/3 \), choosing \( L_3 \) as the independent variable.

Therefore, among any three distinct eigenvalues, the dipole moments should be distributed such that one eigenvalue has zero dipole moment. For sets of any number of distinct eigenvalues, this procedure can be repeated until there are only two remaining eigenvalues. The optimal distribution must have only one or two distinct eigenvalues.

We turn to find the optimal \( L_1, L_2, p_1, \) and \( p_2 \). First we prove an extra condition that will be useful. If \( \xi_r \leq 1/3 \), the optimal distribution cannot have any \( L_i < \xi_r \). This is because we can trivially improve the figure of merit while satisfying the sum rules. Say \( L_1 < \xi_r \) and \( L_2 > 1/3 \). Then we can infinitesimally increase \( L_1 \) and decrease \( L_2 \), such that the sum rule is satisfied, and both moves increase the figure of merit (note that \( L_2 \) can always decrease, as some dipole moment must be distributed to the right of, and not equal to, 1/3. If there were no \( L_i \) greater than 1/3, there could be no \( L_i \) less than 1/3, either). Similarly, if \( \xi_r \geq 1/3 \), then the optimal distribution cannot have any \( L_i \geq \xi_r \). So whatever the
optimal distribution, the position of the eigenvalues relative to \( \xi_r \) must all be the same (\( L_i - \xi_r \geq 0 \) if \( \xi_r < 1/3 \) and \( L_i - \xi_r \leq 0 \) if \( \xi_r > 1/3 \)). In the case of \( \xi_r = 1/3 \) this means that only the single \( L_i = 1/3 \) is optimal.

Rather than write down the optimization problem as a constrained optimization in four variables, we solve the unconstrained problem over just \( L_1 \) and \( L_2 \):

\[
\max_{L_1, L_2} f(L_1, L_2) = \frac{p_1(L_1, L_2)\xi_i}{(L_1 - \xi_r)^2 + \xi_i^2} + \frac{p_2(L_1, L_2)\xi_i}{(L_2 - \xi_r)^2 + \xi_i^2}
\]

where

\[
\begin{align*}
p_1(L_1, L_2) &= \frac{3L_2 - 1}{L_2 - L_1} \\
p_2(L_1, L_2) &= \frac{1 - 3L_1}{L_2 - L_1}
\end{align*}
\]

and \( L_1 \in [0,1/3], L_2 \in [1/3,1] \). The optimal distribution either has \( \partial f/\partial L_1 = 0 \) and \( \partial f/\partial L_2 = 0 \), or lies on the boundary. The derivatives are:

\[
\begin{align*}
\frac{\partial f}{\partial L_1} &= \frac{p_1\xi_i}{x_2 - x_1} \left[ \frac{-2x_1(x_1 - x_2)}{(x_1^2 + \xi_i^2)^2} + \frac{1}{x_1^2 + \xi_i^2} - \frac{1}{x_2^2 + \xi_i^2} \right] \\
\frac{\partial f}{\partial L_2} &= \frac{p_2\xi_i}{x_2 - x_1} \left[ \frac{-2x_2(x_1 - x_2)}{(x_2^2 + \xi_i^2)^2} + \frac{1}{x_2^2 + \xi_i^2} - \frac{1}{x_2^2 + \xi_i^2} \right]
\end{align*}
\]

where \( x_i = L_i - \xi_r \). It will also be useful to have the second derivatives \( \partial^2 f/\partial x_i^2 \) (the mixed derivative won't be necessary), which we write here:

\[
\begin{align*}
\frac{\partial^2 f}{\partial x_1^2} &= \frac{2\xi_i [x_1^2(x_1 + 3x_1) - (3x_1 + x_1)\xi_i^2]}{(x_1^2 + \xi_i^2)^3} \left[ -1 + 3x_2 + 3\xi_r \right] \\
\frac{\partial^2 f}{\partial x_2^2} &= \frac{2\xi_i [x_2^2(x_2 + 3x_1) - (3x_2 + x_1)\xi_i^2]}{(x_2^2 + \xi_i^2)^3} \left[ -1 + 3x_2 + 3\xi_r \right]
\end{align*}
\]

One possible solution for both first derivatives to equal zero is \( x_1 = x_2 \) (i.e. \( L_1 = L_2 = 1/3 \)); however, that is a boundary value we will show later can be ignored (note that the derivative does not blow up if \( x_2 = x_1 \), if you carefully evaluate the two right-hand-side terms). This has the additional benefit that \( p_1 \) and \( p_2 \) can be safely divided out of the problem. For the first derivative to equal zero, we find the simple condition

\[
x_2 = \frac{\xi_i^2 - x_1^2}{2x_1}
\]

The second derivative is identical to the first, but with \( x_2 \leftrightarrow x_1 \), so we also have

\[
x_1 = \frac{\xi_i^2 - x_2^2}{2x_2}
\]

The solutions for both \( x_1 \) and \( x_2 \) cannot be simultaneously satisfied unless \( x_2 = x_1 \), which we have explicitly disallowed. Thus the optimal distribution has at least one eigenvalue on the boundary. We note that if one of the \( L_i = 1/3 \), then both of the \( L_i = 1/3 \), in order for \( \langle L_n \rangle = 1/3 \), further reducing the space of possible values. We now treat four separate cases:

Case 1: \( 0 < \xi_r \leq 1/3 \) In this case we know \( L_1 \geq \xi_r \), which further disallows the boundary value \( L_1 = 0 \). Thus the only possible solutions are \( L_1 = L_2 = 1/3 \), or \( L_2 = 1 \) and \( L_1 \in [\xi_r,1/3) \). If \( L_2 = 1 \), only the single derivative \( \partial f/\partial L_1 \) must equal zero, which previously yielded Eq. (S.10). Solving for \( x_1 \), and discarding the negative solution (\( x_1 \geq 0 \) since \( L_1 \leq \xi_r \)) finds the optimal value \( x_1^* \)

\[
x_1^* = \sqrt{x_2^2 + \xi_r^2} - x_2 = \sqrt{(1 - \xi_r)^2 + \xi_r^2} - 1 + \xi_r
\]

To fulfill the condition \( L_1 \leq 1/3 \) \( (x_1 \leq 1/3 - \xi_r) \), \( \xi_r \) and \( \xi_i \) must satisfy:

\[
\sqrt{(1 - \xi_r)^2 + \xi_r^2} - 1 + \xi_r \leq 1/3 - \xi_r
\]

Solving, one finds

\[
\xi_r^2 \leq 3(\xi_r - 1/3)(\xi_r - 7/9)
\]

which is the equation for a hyperbola in the \((\xi_r, \xi_i)\) plane. Since \( \xi_r \leq 1/3 \), only half of the hyperbola is relevant.

For \((\xi_r, \xi_i)\) that satisfy Eq. (S.14), there remains the question of whether it is a maximum, and if it is a global maximum. We can answer both in the affirmative.

In Eq. (S.9a) for the second derivative with respect to \( x_1 \), we can see immediately that the right-most term in the numerator, \( -1 + 3x_2 + 3\xi_r = -1 + 3L_2 > 0 \). Then by inserting Eq. (S.12) into the term in square brackets in Eq. (S.9a), it is straightforward to show that \( \partial^2 f/\partial x_1^2 < 0 \). Moreover, \( x_1^* \) is the only local optimum for all possible values of \( x_1 \), which means it must also be the global optimum.

Don’t we have to also compare to the boundary value \( L_1 = L_2 = 1/3 \)? Fortunately, no. In the preceding argument, in which \( L_2 \) was fixed at 1, the boundary \( L_1 = 1/3 \) was sufficient, as \( (1/3,1) \) is actually equivalent to \( (1/3,1/3) \) (for \( L_1 = 1/3, p_1 = 3 \) and \( p_2 = 0 \), regardless of the value of \( L_2 \). Thus, \( L_1 = L_2 = 1/3 \) was implicitly checked already, and was found not to be optimal, in the case where \( x_1^* \) was valid (cf. Eq. (S.14)). When there is no interior optimal point, the optimum must occur at \( (1/3,1/3) \), as \( (0,1) \) cannot be optimal (\( L_1 \geq \xi_r \)).

Case 2: \( \xi_r < 0 \) Much of the apparatus of the previous case applies here as well, including the optimal value of \( x_1^* \), Eq. (S.12). Now, however, there is an additional condition on \( \xi_r \), and \( \xi_i \) that restricts \( L_i^* > 0 \) \( (x_1 > -\xi_r) \):

\[
\xi_r^2 \geq 3\xi_r(\xi_r - 2/3)
\]

(S.15)
The same argument as in the preceding section again shows that when \( x_1^* \) is valid, now given by Eqs. \( S.14, S.15 \), it must be the globally optimal point. When it is not valid, however, we must now decide whether \((1/3, 1/3)\) is optimal, or whether \((0, 1)\) is optimal. We can write out

\[
f \left( \frac{1}{3}, \frac{1}{3} \right) - f(0, 1) = \frac{2\xi_i ( -1 + 3\xi_i + 8\xi_r - 9\xi_i^2 )}{9(\xi_i^2 + \xi_r^2)(\xi_i^2 + (1 - \xi_r)^2)(\xi_i^2 + (1/3 - \xi_r)^2)} \tag{S.16}
\]

In the case where Eq. \( S.15 \) is invalidated, it is easy to insert the opposite condition into Eq. \( S.16 \) and verify that the numerator, \( 2\xi_i (2\xi_i - 1) < 0 \), meaning that \((0, 1)\) are the optimal \((L_1, L_2)\). Conversely, when Eq. \( S.14 \) is invalidated, one can verify that \( f(1/3, 1/3) > f(0, 1) \), such that \((1/3, 1/3)\) is optimal.

The next two cases will be highly symmetric with the previous two, so we will just highlight the differences.

**Case 3:** \( 1/3 < \xi_r \leq 1 \)

The difference between this case and the first one is that we now know \( L_1 \leq \xi_r \), disallowing the \( L_1 = 1 \) boundary from being optimal but allowing \( L_1 = 0 \) to potentially be optimal. For \( L_1 = 0 \) \((x_1 = -\xi_r)\), setting \( \partial f / \partial x_2 = 0 \) yields the optimal \( x_2^* \):

\[
x_2^* = -\sqrt{x_1^2 + \xi_i^2} - x_1 = \xi_r - \sqrt{\xi_i^2 + \xi_r^2} \tag{S.17}
\]

where we have taken the negative solution because \( x_1, x_2 \leq 0 \). The condition for \( L_2 \geq 1/3 \) limits the possible \((\xi_r, \xi_i)\):

\[
\xi_i^2 \leq 3 (\xi_r - 1/3) (\xi_r - 1/3) \tag{S.18}
\]

which is half of a hyperbola in the \( \xi \) plane, for \( \xi_r > 1/3 \). By exactly the same arguments as in the first case, when Eq. \( S.18 \) is satisfied, \( x_2^* \) must be globally optimal. When it is not, \((1/3, 1/3)\) must be the optimal value.

**Case 4:** \( \xi_r < 1 \)

Allowing \( \xi_r < 1 \) introduces the extra potentially optimal boundary \( L_2 = 1 \), and it introduces the further condition on \((\xi_r, \xi_i)\) for \( x_2^* \) to be valid:

\[
\xi_i^2 \geq 3 (\xi_r - 1/3) (\xi_r - 1) \tag{S.19}
\]

If this further condition is met, then \( x_2^* \) is globally optimal. Otherwise, we again compare \( f(1/3, 1/3) \) to \( f(0, 1) \), through Eq. \( S.16 \), when the conditions are not met. One finds that when Eq. \( S.19 \) is not met, \((0, 1)\) is optimal, and when Eq. \( S.18 \) is not met, \((1/3, 1/3)\) is optimal.

We can collate the results of the four cases into a single tedious but exact analytical representation for the optimal depolarization factors, \((L_1, L_2)_{\text{opt}}\), for any possible material:

\[
(L_1, L_2)_{\text{opt}} = \begin{cases}
(0, 1) & \xi_r < 0; \xi_i^2 < 3\xi_i \left( \xi_r - \frac{2}{3} \right) \text{ or,} \\
(\xi_r + \Delta_1, 1) & \xi_r > 1; \xi_i^2 < 3 \left( \xi_r - \frac{1}{3} \right) \left( \xi_r - \frac{2}{3} \right) \\
(0, \xi_r - \Delta_2) & \frac{1}{3} < \xi_r < 1; \xi_i^2 < 3 \left( \xi_r - \frac{1}{3} \right) \left( \xi_r - \frac{2}{3} \right) \text{ or,} \\
(\frac{1}{3}, 1) & \xi_r < \frac{1}{3}; \xi_i^2 > 3 \left( \xi_r - \frac{2}{3} \right) \left( \xi_r - \frac{1}{3} \right) \text{ or,} \\
& \xi_r > \frac{1}{3}; \xi_i^2 > 3 \left( \xi_r - \frac{1}{3} \right) \left( \xi_r - \frac{2}{3} \right)
\end{cases} \tag{S.20}
\]

where

\[
\Delta_1 = (1 - \xi_r) \left( \sqrt{1 + \frac{\xi_i^2}{(1 - \xi_r)^2}} - 1 \right) \tag{S.21a}
\]

\[
\Delta_2 = \xi_r \left( \sqrt{1 + \frac{\xi_i^2}{\xi_r^2}} - 1 \right) \tag{S.21b}
\]

The optimal depolarization factors are mapped out in Fig. \( S.1 \) which carefully delineates each of the regions. \( \Delta_1 = \Delta_2 = 0 \) in the case where \( \xi_i = 0 \), yielding the \((\xi_r, 1)\) or \((0, \xi_r)\) optimal values that are used in the main text. We can see why those values, although not exactly correct when \( \xi_i > 0 \), are very accurate in most cases. Because the optimal eigenvalues must lie on the boundary, one of the optimal values will usually be exactly correct (e.g. \( L_2 \) in the case \( 0 \leq \xi_i \leq 1/3 \)), with the other value off only by a small factor given by Eq. \( S.21 \). Fig. \( S.2 \) shows the very small error between Eq. (7) and the exact bounds for most materials.
FIG. S1. Demarcation of the optimal depolarization factors in the \((\xi_r, \xi_i)\) plane. The solution presented in the main text is the exact solution for \(\xi_i = 0\).

FIG. S2. Image: fundamental limits to \(\sigma_{\text{ext}} \lambda / V\) as a function of \(\xi\) in the complex plane. Contour lines: relative error \(\delta\) between simplified solution, Eq. (7) in the main text, and the exact solution, Eq. (S.20). For most materials, the error is very small, and the simplified solution is sufficient.

One can see from Fig. S1 that the space of theoretically possible materials is largely covered by either the solution \((0, 1)\) or the solution \((1/3, 1/3)\), for which either flat disks or spheres reach the optimum. There is an additional curve along which the optimal solution is \((0, 1/2)\), for which the cylinder is optimal. Thus we see why spheroids perform so well. One should note, as seen in Fig. S2, that for most materials \(0 < \xi_r < 1/3\) and \(\xi_i < 0.1\), for which spheroids do not reach the upper bound and some improvement beyond spheroids is possible.

SHORT PROOFS OF THE SUM RULES

Uncoated particles

We provide a unified framework to derive both sum rules, utilizing work by Ouyang and Isaacson [1] and Fuchs [2], while also introducing the concept of “resolution of the identity,” which is well-known in quantum mechanics but appears to not have been recognized explicitly in previous work on integral equation formulations of the scattering problem. The derivation here of the second sum rule avoids the continuous-to-discrete-to-continuous transformations of [2], potentially simplifying the proof.

Following [1] we define the linear operator

\[
B[\sigma](x) = \int_{S} \frac{1}{|x - x'|} \sigma(x') d^2x'
\]

which is positive definite and symmetric, such that it has a unique Cholesky decomposition

\[
B = U^T U
\]

where \(U\) is upper triangular. So-called “Plemelj symmetrization” yields

\[
K^T B = BK
\]

showing that \(BK\) is symmetric. We define another symmetric operator

\[
D = (U^{-1})^T BK U^{-1} = UKU^{-1}
\]

which has the same eigenvalue spectrum as \(K\). Defining \(s_n\) to be the eigenvectors of \(D\) \((Ds_n = \lambda_n s_n)\), there is a simple relationship between \(s_n\) and \(\sigma_n\):

\[
\sigma_n = U^{-1} s_n
\]

\(D\) is complete, leading to the resolution of the identity...
we need:

\[
\delta(x - x') = \sum_n s_n(x') s_n(x) = \sum_n \sigma_n(x') U^T U \sigma_n(x) = \sum_n \sigma_n(x') B \sigma_n(x) = \sum_n \tau_n(x') \sigma_n(x) \tag{S.26}
\]

This identity yields the two sum rules. For the first, we simply sum over the dipole strengths of every mode

\[
\sum_{n} p_{n, \alpha \beta} = \frac{1}{V} \int_S \int_{S'} n_{\alpha}(x) \sum_n \tau_n(x') \sigma_n(x') x'_\beta d^2 x' d^2 y = \int_S d^2 y K(x, y) \sum_n \tau_n(x') \sigma_n(y) = K(x, x') \tag{S.27}
\]

For the second sum rule, we use the crucial formula

\[
\sum_n \tau_n(x') \lambda_n \sigma_n(x) = K(x, x') \tag{S.28}
\]

which can be proven by taking

\[
\sum_n \tau_n(x') \lambda_n \sigma_n(x) = \sum_n \tau_n(x') \int_S K(x, y) \sigma_n(y) d^2 y = \int_S d^2 y K(x, y) \sum_n \tau_n(x') \sigma_n(y) = K(x, x') \tag{S.29}
\]

where in the final step we have again used the resolution of the identity. Now we can write

\[
\sum_n \lambda_n p_{n, \alpha \alpha} = \frac{1}{V} \sum_{\alpha} \int_S d^2 x \int_S d^2 x' n_{\alpha}(x) x'_\alpha \sum_n \tau_n(x') \lambda_n \sigma_n(x') = \frac{1}{V} \int_S d^2 x \int_S d^2 x' \sum_{\alpha} x'_\alpha K(x', x) n_{\alpha}(x) = \frac{1}{V} \int_S d^2 x \int_S x \cdot n(x') \int_{V_m} \nabla \cdot x_{\alpha} d^3 x = \delta_{\alpha \beta} \frac{1}{V} \int_{V} \nabla \cdot x_{\alpha} d^3 x \tag{S.30}
\]

The final integral in Eqn. [S.30] is worked out in [2] and shown to be equal to $-1/2$, for either a single particle or a collection of particles. Thus we have

\[
\sum_{n, \alpha} \lambda_n p_{n, \alpha \alpha} = -\frac{1}{2} \tag{S.31}
\]

or, in terms of the depolarization factor

\[
\langle L_n \rangle = \frac{\sum_{n, \alpha} L_{n} p_{n, \alpha \alpha}}{\sum_{n, \alpha} p_{n, \alpha \alpha}} = \frac{1}{3} \tag{S.32}
\]

Coated particles

Here we treat the coated particle case, partly utilizing an idea from Ref. [3]. The integral equation remains the same except that $\Lambda \to \hat{\Lambda}$, where $\Lambda$ is a diagonal matrix operator. If we restrict ourselves to only two materials, we can write $\hat{\Lambda} = \Lambda C$, where:

\[
\hat{C} = \begin{pmatrix}
\hat{I} & \cdots \\
-\hat{I} & \ddots
\end{pmatrix} \tag{S.33}
\]

and $\Lambda = (\epsilon_{\text{int}} + \epsilon_{\text{ext}}) / 2 (\epsilon_{\text{int}} - \epsilon_{\text{ext}})$. The integral equation is then:

\[
\hat{C} \Lambda \sigma - \hat{K} \sigma = s \tag{S.34}
\]

which is a generalized eigenvalue equation. Since $C$ is real and symmetric, the orthogonality condition becomes:

\[
\langle \sigma_n, \hat{C} \tau_m \rangle = \delta_{mn} \tag{S.35}
\]

which yields an altered resolution of the identity:

\[
\delta(x, x') = \sum_n \tau_n(x') C(x) \sigma_n(x) = \sum_n C(x') \tau_n(x') \sigma_n(x) \tag{S.36}
\]

Now when we sum over the dipole strengths we find:

\[
\sum_n p_{n, \alpha \beta} = \frac{1}{V} \int_S \int_{S'} n_{\alpha}(x) C(x) \sum_n \tau_n(x') C(x) \sigma_n(x') x'_\beta d^2 x' d^2 x' = \frac{1}{V} \int_S n_{\alpha} C(x) x'_\beta d^2 x = \delta_{\alpha \beta} \frac{1}{V} \sum_m (-1)^m \int_{V_m} \nabla \cdot x_{\alpha} d^3 x \tag{S.38}
\]

where $C$ contributed opposite signs to the interior and exterior regions, alternately adding and subtracting concentrically larger volumes. Ultimately, the sum rule is reduced by the volume fraction of the “interior” material, typically metallic. The same reasoning yields a similar term in the sum $\sum_{n} p_n L_n$, such that both the numerator and denominator of the second sum rule contain the volume fraction, and the ratio of the two remains the same as for the uncoated–particle case.