Sum rules for Mie scattering

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By making use of the recently derived asymptotic expansion to the Mie forward-scattering amplitude, we are able to employ complex contour-integration techniques to derive three sum rules in the case of a constant real refractive index. The results are in good agreement with numerical calculation. Extension to complex refractive indices may be possible under certain analyticity conditions.

INTRODUCTION

The Mie theory forward-scattering amplitude S is an analytic function of the size parameter $x = kr$, where $k = 2\pi/\lambda$, r is the particle radius, and λ is the wavelength.[1,2] In the sign convention that writes the electromagnetic plane wave in the form $\exp[i(\omega t - kz)]$, the region in which S is analytic is the lower half of the complex x (or complex k) plane.[1–4] This analyticity region is determined ultimately by causality, the fact that scattering cannot occur before the incident wave arrives at the scatterer.[5] $S(x)$ also obeys a crossing relation, which for real values of x is $S(-x) = S^*(x)$, and is derived from the requirement of reality of the electric field.[2,3] For complex values of x this argument gives

$$S(x) = S^*(-x^*). \tag{1}$$

As a particular case, we note that $S(x)$ is purely real on the negative imaginary axis.

In a recent paper,[3] we used this crossing condition to generalize the asymptotic approximation for the Mie extinction efficiency obtained recently by Nussenzveig and Wiscombe.[6] We obtained

$$\begin{aligned}\frac{4}{x^2}S(x) = {} & 2 + 1.9923861(1 - i\sqrt{3})x^{-2/3} \\ & + i\frac{8}{x}\left\{\frac{m^2+1}{4\sqrt{m^2-1}} - F\exp[-2i(m-1)x]\right. \\ & -\sum_{j=1}^{\infty}\left(\frac{m-1}{2j+1-m}\right)\left(\frac{m-1}{m+1}\right)^{2j} \\ & \left.\times \exp[-2i(m-1+2jm)x]\right\} \\ & - 0.7153537(1 + i\sqrt{3})x^{-4/3} \\ & - i0.166(1 - i\sqrt{3})\frac{m^2+1}{(m^2-1)^{3/2}} \\ & \times (2m^4 - 6m^2 + 3)x^{-5/3} \\ & + 0(x^{-2}) + \text{ripple},\end{aligned} \tag{2}$$

where

$$F = \frac{m^2}{(m+1)(m^2-1)}\left[1 - \frac{i}{2x}\left(\frac{1}{m-1} - \frac{m-1}{m}\right)\right]$$

and $m = n - i\kappa$ is the complex refractive index.

The result of Nussenzveig and Wiscombe can be readily obtained from this expression by using the relation[3]

$$Q_{\text{ext}} = \frac{4}{x^2}\,\text{Re}\,S. \tag{3}$$

The difference between the exact value of $4S(x)/x^2$ and its approximation, as represented by the explicit terms on the right-hand side of Eq. (2), is analytic in the lower half-plane, except at $x = 0$, and is of order $0(X^{-2})$ as $|X| \to \infty$. Moreover, both $4S(x)/x^2$ and its approximation are order $0[\exp(|x|^{\beta})]$ for $0 < \beta < 1$ (the first from the Kramers–Kronig relation and the second by inspection), so the difference is also of this order. The Phragmén–Lindelöf theorem[7] then guarantees that the difference is $0(X^{-2})$ on the contour $X = |X|$ in the lower half-plane. Thus the asymptotic approximation (2) is valid in the entire lower half-plane for sufficiently large $|X|$ and not just on the real axis. Since Eq. (2) was obtained by using complex-variable techniques, its validity in the complex plane is not unexpected. (See also Ref. 8.)

Having such an asymptotic expansion in our possession, we may now use contour-integration techniques to derive a series of sum rules of the form (see Fig. 1)

$$\int_{X_1}^{X_2} S(x)f(x)\mathrm{d}x = F(X_1, X_2).$$

Obvious special cases are $X_1 = 0$ and $X_2 = \infty$.

Note that, in practice, our assumptions break down for very large $|k|$, since above the plasma frequency $m(k) \to 1$, and there is no scattering. Furthermore, above the pair-production threshold, quantum electrodynamic effects become important. For this reason, we avoid setting $X_2 = \infty$ in this paper.

Before we consider letting $X_1 = 0$, however, we must note the behavior of $S(x)$ as $x \to 0$. Since our sign convention for

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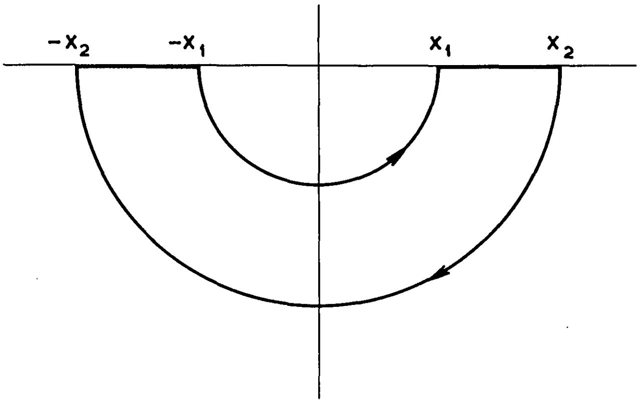

Fig. 1. Integration contour for general sum rules.

the phase of the exponential is the opposite of that of Bohren and Huffman, the relevant result is[1,9]

$$\lim_{x\to 0} S(x) = ix^3\frac{m^2-1}{m^2+2}. \tag{4}$$

One final consideration before actually tackling any contour integrals is the refractive index. In general, of course, m is a function of k and hence of x for fixed r,[1,2,10] and much is known about the analytical properties of $m(k)$. From the corresponding properties of $\epsilon(k)$ and $\mu(k)$ (Chap. 7 of Ref. 10 and Sec. 82 of Ref. 11) it follows immediately that

(i) m is an analytic function of k in the lower half-plane,
(ii) m obeys the crossing relation (1) as a function of k.

At fixed r, we may translate these properties into the same properties for $m(x)$. While only information about the asymptotic properties of $m(k)$ is then required to obtain the Kramers–Kronig relations[1,4,10,11] m, we will not need to use the relations but will utilize the properties (i) and (ii) above, since these are necessary to obtain the equivalent properties of S when m is a function of k. While these conditions are quite mild, they are restrictive.

Note that it is possible to construct models of $m(k)$ that satisfy conditions (i) and (ii) without satisfying the Kramers–Kronig relations, because the asymptotic behavior is different. An example is a constant real value $m(k) = m_0 \neq 1$. In practice, we need not take the contour at large x (or large k) to frequencies above the plasma frequency, at which $m(k) \to 1$, so it is reasonable to take $m_0 \neq 1$. Moreover, in performing the integrals around the contours $|X| = X_1$ and $|X| = X_2$, we need only the value of m on the contours. We will, in general, assume that $m = m_1$ on the contour $|X| = X_1$ and $m = m_2$ on the contour $|X| = X_2$, where m_1 and m_2 are real constants that will in general be different. We note from the approximate formula for the dielectric constant given by Jackson, Eq. (7.51) of Ref. 10, that the approximation $n(k)$ = real constant is good for k far from the resonant poles. Note that poles can occur between X_1 and X_2 as long as X_1 and X_2 themselves are far from the poles, and in this case the constants m_1 and m_2 will be different (Fig. 1).

THREE-WEIGHTED SUM RULES

In this section, we derive in full detail three sum rules. We take $X_1 = 0$, m real and constant, and $f(x)$ a negative power of x. Consider now the contour in Fig. 2. The function $S(x)x^{-2}$ has no poles within that contour, and so from Cauchy's theorem[12]

$$\int_{C_1}\frac{4}{x^2}S(x)\mathrm{d}x = 0,$$

$$\therefore \int_{-X}^{X}\frac{4}{x^2}S(x)\mathrm{d}x = \int_{C}\frac{4}{x^2}S(x)\mathrm{d}x, \tag{5}$$

where C is the curved part of the contour C_1 traversed in an anticlockwise direction. Now, from Eq. (1), we see that the imaginary part of the integrand on the left-hand side of Eqs. (5) is odd, whereas the real part is even, so Eqs. (5) reduce to

$$\int_0^X Q(x)\mathrm{d}x = \frac{1}{2}\,\mathrm{Re}\int_C\frac{4}{x^2}S(x)\mathrm{d}x$$

$$= \frac{1}{2}\,\mathrm{Re}\int_{-\pi}^{0} 4X^{-2}e^{-2i\phi}S(Xe^{i\phi})iXe^{i\phi}\mathrm{d}\phi. \tag{6}$$

When Eq. (2) is inserted into Eq. (6), the result becomes

$$\begin{aligned}\int_0^X Q(x)\mathrm{d}x = {} & 2X + 3 * 1.9923861X^{1/3} - \pi\frac{m^2+1}{\sqrt{m^2-1}} \\ & - 8\frac{m(m-1)}{(m+1)^2}\left\{Si[2(m-1)X] - \frac{\pi}{2}\right\} \\ & + 4\frac{m^2 - m(m-1)^2}{(m^2-1)^2}\cos[2(m-1)X]X^{-1} \\ & - 8\sum_{j=1}^{\infty}\left(\frac{m-1}{2j+1-m}\right)\left(\frac{m-1}{m+1}\right)^{2j} \\ & \times\left\{Si[2(m-1+2jm)X] - \frac{\pi}{2}\right\} \\ & + 3 * 0.7153537X^{-1/3} + \frac{3\sqrt{3}}{2} * 0.166 \\ & \times\frac{m^2+1}{(m^2-1)^{3/2}}(2m^4 - 6m^2 + 3)X^{-2/3} + 0(X^{-1}),\end{aligned} \tag{7}$$

where

$$Si(z) = \int_0^z t^{-1}\sin t\mathrm{d}t \tag{8}$$

is the sine integral.[13]

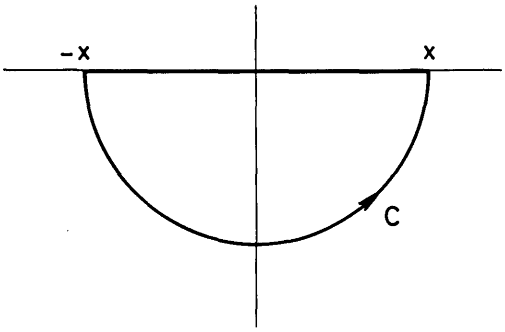

Fig. 2. Integration contour C_1 for the three-weighted sum rules.

Our second result is obtained by considering the integrand $S(x)x^{-3}$. In this case, the real part is odd and the imaginary part even, so by using similar reasoning to the above we find that

$$\begin{aligned}
\int_0^X \frac{4}{x^3}\,\mathrm{Im}\,S(x)\mathrm{d}x &= \frac{1}{2}\,\mathrm{Im}\int_C \frac{4}{x^3}S(x)\mathrm{d}x \\
&= \pi + \frac{3\sqrt{3}}{2} * 1.9923861X^{-2/3} \\
&\quad - 2\frac{m^2+1}{\sqrt{m^2-1}}X^{-1} + 8\frac{m(m^2-m+1)}{(m+1)^2} \\
&\quad \times \left\{Si[2(m-1)X] - \frac{\pi}{2}\right\} \\
&\quad + 4\frac{m(m^2-m+1)}{(m+1)(m^2-1)}\cos[2(m-1)X]X^{-1} \\
&\quad - 2\frac{m^2-m(m-1)^2}{(m^2-1)^2}\sin[2(m-1)X]X^{-2} \\
&\quad + 8\sum_{j=1}^{\infty}\left(\frac{m-1}{2j+1-m}\right)\left(\frac{m-1}{m+1}\right)^{2j} \\
&\quad \times \Bigg(\cos[2(m-1+2jm)X] \\
&\quad + 2(m-1+2jm)X \\
&\quad \times \left\{Si[2(m-1+2jm)X] - \frac{\pi}{2}\right\}\Bigg)X^{-1} \\
&\quad + \frac{3\sqrt{3}}{4} * 0.7153537X^{-4/3} \\
&\quad + \frac{3}{5} * 0.166\frac{m^2+1}{(m^2-1)^{3/2}} \\
&\quad \times (2m^4 - 6m^2 + 3)X^{-5/3} + 0(X^{-2}). \qquad (9)
\end{aligned}$$

Our third result is obtained by considering the integrand $S(x)x^{-4}$, with the real part even and the imaginary part odd. From Eq. (4) we see that we now have a simple pole at the origin. This can be handled either by distorting the contour of Fig. 2 by using a small semicircle around the origin or by using Cauchy's integral formula in the form[12]

$$\frac{1}{2}S(x)x^{-3} = \frac{1}{2\pi i}P\int_{C_1}\frac{S(z)z^{-3}}{z-x}\mathrm{d}z, \qquad (10)$$

where x is a point on the contour C_1 and P stands for the Cauchy principal value. When the limit $x \rightarrow 0$ is taken, we obtain

$$\begin{aligned}
\int_0^X \frac{4}{x^4}\,\mathrm{Re}\,S(x)\mathrm{d}x &= \int_0^X x^{-2}Q(x)\mathrm{d}x \\
&= \frac{1}{2}\,\mathrm{Re}\int_C \frac{4}{x^4}S(x)\mathrm{d}x - \text{pole term} \\
&= 2\pi\frac{m^2(0)-1}{m^2(0)+2} \\
&\quad - 2X^{-1} - \frac{3}{5} * 1.9923861X^{-5/3} \\
&\quad + \frac{16}{3}\frac{m(m-1)^2}{m+1}\left\{Si[2(m-1)X] - \frac{\pi}{2}\right\} \\
&\quad + \frac{4}{3}\frac{m}{m+1}\sin[2(m-1)X]X^{-2} \\
&\quad + \frac{8}{3}\frac{m(m-1)}{m+1}\cos[2(m-1)X]X^{-1} \\
&\quad + \frac{4}{3}\frac{m^2-m(m-1)^2}{(m^2-1)^2} \\
&\quad \times \cos[2(m-1)X]X^{-3} \\
&\quad + 4\sum_{j=1}^{\infty}\left(\frac{m-1}{2j+1-m}\right)\left(\frac{m-1}{m+1}\right)^{2j} \\
&\quad \times \Bigg(\sin[2(m-1+2jm)X] \\
&\quad + 2(m-1+2jm)X \\
&\quad \times \cos[2(m-1+2jm)X] \\
&\quad + 4(m-1+2jm)^2X^2 \\
&\quad \times \left\{Si[2(m-1+2jm)X] - \frac{\pi}{2}\right\}\Bigg)X^{-2} \\
&\quad + \frac{3}{7} * 0.7153537X^{-7/3} \\
&\quad + \frac{3\sqrt{3}}{8} * 0.166\frac{m^2+1}{(m^2-1)^{3/2}} \\
&\quad \times (2m^4 - 6m^2 + 3)X^{-8/3} + 0(X^{-3}). \qquad (11)
\end{aligned}$$

In this equation the first term on the right-hand side is derived from the pole at $x = 0$, and so the refractive index m is to be evaluated in the static limit at $k = 0$; hence the notation $m(0)$ in this term. The value of m that occurs in the other terms on the right is derived from the integral around the contour and is to be evaluated at $k = X/r$.

The contribution from the oscillating terms, $Si(z)$, $\cos(z)$, and $\sin(z)$, in Eqs. (7), (9), and (11) is in fact quite small, as can be seen from a quick glance at the asymptotic expansion of the sine integral[13]

$$Si(z) \sim \frac{\pi}{2} - \cos(z)/z - \sin(z)/z^2 + \ldots. \qquad (12)$$

In fact, in the majority of cases, the first two or three terms should usually be sufficient for a reasonably accurate result.

NUMERICAL VERIFICATION

We have checked our three sum rules numerically, for real refractive indices of 1.1 and 1.5. Calculations for a total of 40 values of X between 200 and 400 were performed, with an integration step size of $\Delta x = 0.025$.

For the first sum rule [Eq. (7)] the maximum discrepancy between the numerical and analytic results was 0.05% for a refractive index of 1.5 and 0.07% for a refractive index of 1.1. We also varied the coefficients of the various terms in Eq. (7) to examine their sensitivity. The coefficient of the first term, namely, 2, could be varied by no more than 0.1% before serious disagreement with the numerical results was noted. The second term could be varied by a few percent and the third by about 20% before serious disagreement was noted. All other terms could be varied by 100% without a significant effect on the accuracy. For a refractive index of 1.1, the same sensitivity was observed.

For the second sum rule [Eq. (9)] the average discrepancy for each refractive index was larger by a factor of 3. The first term, π, could again be varied by no more than 0.1% and the second term by a few percent, whereas all other terms could be varied by more than 100% before the accuracy was affected.

In the case of the third sum rule [Eq. (11)] the result is totally dominated by the first (pole) term, with the consequence that all other terms could be varied by at least 100% before any change was observed. The difference between the numerical and analytical results—roughly 1 part in 10^5—is almost certainly due to numerical quadrature errors.

We may thus conclude that the leading term in each of our three sum rules has been verified with high accuracy, while the term involving 1.9923861 has twice been confirmed to within a few percent. Most other terms make such a small contribution (at least for $X > 200$) that they may be neglected.

In a less comprehensive series of calculations, we have also examined the accuracy of these sum rules for smaller values of X. In the case of a refractive index of 1.5, all three remain quite accurate (no more than 1% discrepancy) down to at least $X = 20$, provided that sufficient terms are included.

For a refractive index of 1.1, all three sum rules are accurate down to $X = 50$, and the last two sum rules are quite accurate down to about $X = 25$. For refractive indices near unity, the term derived from

$$0.166 \frac{m^2 + 1}{(m^2 - 1)^{3/2}} (2m^4 - 6m^2 + 3)$$

becomes significant in each sum rule, especially for small X. We have been able to improve significantly the accuracy of the first sum rule [Eq. (7)] for X less than about 300, by doubling the contribution of this term. Some improvement was also noted in the other two sum rules for small values of X. We would therefore like to suggest tentatively that this particular term may in fact be too small by a factor of perhaps 2.

APPLICATIONS

So far, the only weighting functions that we have employed have been simple powers. One obvious extension of these ideas leads us to two sum rules for the extinction coefficient, β.[4,9] Consider first

$$\begin{aligned} K^{-1} \int_0^K \beta(k)\mathrm{d}k &= K^{-1} \int_0^K \mathrm{d}k \int_0^\infty \pi r^2 n(r) Q(kr)\mathrm{d}r \\ &= \int_0^\infty \mathrm{d}r \pi r n(r) K^{-1} \int_0^{Kr} Q(x)\mathrm{d}x \\ &= \int_0^\infty \mathrm{d}r \pi r n(r) K^{-1} \\ &\quad \times \left[2Kr + 3 * 1.9923861 (Kr)^{1/3} \right. \\ &\quad \left. - \pi \frac{m^2 + 1}{\sqrt{m^2 - 1}} + 3 * 0.7153537 (Kr)^{-1/3} \ldots \right] \\ &= 2\pi M_2 + 3 * 1.9923861 \pi M_{4/3} K^{-2/3} \\ &\quad - \pi^2 \frac{m^2 + 1}{\sqrt{m^2 - 1}} M_1 K^{-1} \\ &\quad + 3 * 0.7153537 \pi M_{2/3} K^{-4/3} \\ &\quad + \text{higher-order items.} \end{aligned} \qquad (13)$$

Similarly, we may consider

$$\begin{aligned} K \int_0^K k^{-2} \beta(k)\mathrm{d}k &= \int_0^\infty \mathrm{d}r \pi r^3 n(r) K \int_0^{Kr} Q(x) x^{-2}\mathrm{d}x \\ &= 2\pi^2 \frac{m^2(0) - 1}{m^2(0) + 2} M_3 K - 2\pi M_2 \\ &\quad - 0.6 * 1.9923861 \pi M_{4/3} K^{-2/3} + \ldots . \end{aligned} \qquad (14)$$

Here

$$M_\nu = \int_0^\infty r^\nu n(r)\mathrm{d}r,$$

and $n(r)\mathrm{d}r$ is the number of spherical particles per unit volume with radii between r and $r + \mathrm{d}r$. [Alternatively, we may interpret $n(r)$ as the columnar size distribution and replace β by the optical depth τ.]

In a recent paper,[14] Viera and Box showed that knowledge of M_2 could prove extremely valuable in the inversion of multispectral extinction data to obtain the size distribution $n(r)$. By performing the integrals indicated in Eqs. (13) and (14) for a series of cutoffs K, it may be possible to obtain a reasonably reliable estimate of M_2.

Our middle sum rule [Eq. (9)] is clearly less valuable in this context, as measurements of Im S cannot be performed directly. However, it may be possible to perform a Kramers–Kronig analysis[1,8,15] in order to obtain this function. Some improvement in noise level may result from the averaging nature of the integrations involved.

One further application is illuminating. If we define Q_{NW} to be given by the real part of Eq. (2), we may then integrate the first two terms of this expression from 0 to X, and we note that they agree with the corresponding terms in Eq. (7).

We have already noted that these terms dominate for large X. It is probably better to avoid the pole at the origin in Eq. (2) and instead concentrate on integrals from X_1 to X_2. In this case, the third term in Eq. (7) drops away, and we note that

$$\int_{X_1}^{X_2} Q\mathrm{d}x = \int_{X_1}^{X_2} Q_{\mathrm{NW}}\mathrm{d}x + 0({X_1}^{-1}),$$

at least for real m, and X_1 is at least 10. From this result we may conclude that Q_{NW} is not only an asymptotic approximation to Q but is also an approximation in the mean to Q. Q_{NW} lacks the fine ripple structure of Q, but it will reproduce the general shape of the function, on the average. This property of Q_{NW} may be usefully exploited in approximate calculation of the extinction coefficient for polydispensions, in which the size distribution tends to smear out the ripple structure.

DISCUSSION

Sum rules for various optical properties are not new,[16–20] and none of these three is entirely new. In an earlier paper,[4] two of us obtained, by using similar methods, a result that is closely related to Eq. (7). Translated into the notation of the present paper (and converted from a polydispension to a monodispension), this result may be written as

$$X^{-1}\int_0^X Q(x)\mathrm{d}x = 2 + 3\,(\sqrt{3}a_R + a_I)X^{-2/3},$$

where a_R and a_I are empirical constants derived by van de Hulst.[9] Although the numerical value of the coefficient of $X^{-2/3}$ differs somewhat from the coefficient in (more recent) the asymptotic approximation of Nussenzweig and Wiscombe,[3] the functional form is seen to be correct.

In the same paper,[4] we also obtained, from a different approach, the $X \rightarrow \infty$ limit of Eq. (9) (again applied to a polydispension rather than to a monodispension). The $X \rightarrow \infty$ limit of Eq. (11) was originally obtained by Purcell,[18] in the more general case of spheroidal scatterers. We emphasize again that since the term $[m^2(0) - 1]/[m^2(0) + 2]$ comes from the pole at the origin, it is to be evaluated at $k = 0$, i.e., $m^2(0)$ is the static dielectric constant.

Although such infinite-range sum rules have the advantage that they may provide informative hints about the physics above the (usually enforced) cutoff limits, finite-range sum rules (also known as finite-energy sum rules[21]) may be even more useful in certain circumstances, as we have seen in the previous section.

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