Anomalous diffraction of light with geometrical path statistics of rays and a Gaussian ray approximation

M. Xu, M. Lax, and R. R. Alfano

Institute for Ultrafast Spectroscopy and Lasers, New York State Center of Advanced Technology for Ultrafast Photonic Materials and Applications and Department of Physics, The City College and Graduate Center of City University of New York, New York, New York 10031

Received August 13, 2002

The anomalous-diffraction theory (ADT) of extinction of light by soft particles is shown to be determined by a statistical distribution of the geometrical paths of individual rays inside the particles. Light extinction depends on the mean and the mean-squared geometrical paths of the rays. Analytical formulas for optical efficiencies from a Gaussian distribution of the geometrical paths of rays are derived. This Gaussian ray approximation reduces to the exact ADT in the intermediate case of light scattering for an arbitrary soft particle and describes well the extinction of light from a system of randomly oriented and (or) polydisperse particles. The implications for probing of the sizes and shapes of particles by light extinction are discussed. © 2003 Optical Society of America

OCIS codes: 290.2200, 290.5850, 290.4020, 280.1310.

Anomalous-diffraction theory (ADT) which was introduced by van de Hulst[1] for light extinction and scattering, is one of the simplest and most powerful approximations of the interaction of electromagnetic radiation with spherical and nonspherical soft particles. This approach has been used in remote sensing of cirrus clouds and climate research, in biophysical and biomedical research, and in other applications.[2] The anomalous-diffraction theory is based on the premise that the extinction of light by a particle is primarily a result of the interference between the rays that pass through the particle with those that do not.[3] This approximation is most applicable to so-called soft particles with the complex relative refractive index m near 1 ($|m-1| \ll 1$) and with a characteristic dimension of size r exceeding wavelength λ of the incident radiation ($2\pi r/\lambda > 1$) to achieve a high degree of accuracy.[3–6] This accuracy has been observed to improve with softness and nonsphericity,[6] and with polydispersity of the particle.[3]

In this Letter we show that ADT has a statistical interpretation. The extinction of light by particles measures a probability distribution of the geometrical path of the individual rays inside the particles rather than the sizes and shapes of individual particles.

In the framework of ADT,[1] the extinction, absorption, and scattering efficiencies of a particle are given by

$$\begin{aligned} Q_{\text{ext}} &= \frac{2}{P}\,\Re \iint_P \{1-\exp[-ikl(m_r-1)] \\ &\quad \times \exp(-klm_i)\}\mathrm{d}P, \\ Q_{\text{abs}} &= \frac{1}{P}\iint_P [1-\exp(-2klm_i)]\mathrm{d}P, \\ Q_{\text{sca}} &= Q_{\text{ext}} - Q_{\text{abs}}, \end{aligned} \tag{1}$$

where $\Re$ represents the real part, the wave number is $k = 2\pi/\lambda$ for wavelength λ, the complex relative refractive index is $m = m_r - im_i$, l is the geometrical path of an individual ray inside the particle, and P is the projected area of the particle in the plane perpendicular to the incident light over which the integration is performed. The optical efficiencies for a system of randomly oriented and (or) polydisperse particles are averaged over all the sizes and orientations of particles weighted by their projection areas, i.e.,

$$\overline{Q} = \frac{\sum PQ}{\sum P}. \tag{2}$$

The integration in Eq. (1) over the projected area for a single particle at a fixed orientation or the averaging in Eq. (2) over the combined projected area from all sizes and orientations of particles can be reinterpreted as an averaging over a distribution of the geometrical path l of rays. By dividing the (combined) projection area into equal-area elements and counting the resultant geometrical paths that correspond to each projection area element according to their lengths, one can find a probability function $p(l)\mathrm{d}l$ that describes the probability that geometrical path l from a ray is within $(l, l+\mathrm{d}l)$. The probability function is normalized to $\int p(l)\mathrm{d}l = 1$. By this interpretation, we can rewrite the optical efficiencies in Eq. (1) as expected values in accordance with probability distribution $p(l)$ of the geometrical paths of rays. The extinction and absorption efficiencies in Eq. (1) can be expressed as

$$\begin{aligned} Q_{\text{ext}} &= 2\Re\int \{1-\exp[-ikl(m_r-1)] \\ &\quad \times \exp(-klm_i)\}p(l)\,\mathrm{d}l, \\ Q_{\text{abs}} &= \int [1-\exp(-2klm_i)]p(l)\,\mathrm{d}l. \end{aligned} \tag{3}$$

Assume that the geometrical path distribution of rays (in short, the ray distribution) for one particle with a unit size is $p_0(l)$; then the ray distribution for a particle with the same shape, orientation, and a different size L is given by $p(l) = (1/L)p_0(l/L)$ from scaling of

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length. Thus, a system of such particles whose size is distributed according to a probability-density function $n(x)$ has a ray distribution function

$$p_{\mathrm{pol}}(l) = \frac{\int (1/x) p_0(l/x) n(x) x^2 \,\mathrm{d}x}{\int n(x) x^2 \,\mathrm{d}x}, \tag{4}$$

weighted by the projection area of individual particles that is proportional to x^2. The subscript pol or rn is used to denote a polydispersed particle or one that is randomly oriented, respectively.

Let us consider a unit spheroid with a semisize $b = 1$ of the revolutional axis and an axial ratio ϵ and with an angle χ between the propagation direction of the incident beam and the revolutional axis of the spheroid. The geometrical length of a ray and the projection area for such a spheroid have been calculated.[5] The geometrical path distribution of the rays can then be found:

$$p_0(l) = \frac{1}{2}(\epsilon^{-2}\sin^2\chi + \cos^2\chi) l H(l) \times H\left[\frac{2}{(\epsilon^{-2}\sin^2\chi + \cos^2\chi)^{1/2}} - l\right], \tag{5}$$

where $H(x)$ is a Heaviside function. The ray distribution for a system of such spheroids at a fixed orientation χ with a log-normal size distribution,[7]

$$n(x) = \frac{1}{(2\pi)^{1/2}\sigma} r^{-1} \exp\left[-\frac{\ln^2(r/a_m)}{2\sigma^2}\right], \tag{6}$$

is given by

$$p_{\mathrm{pol}}(l) = \frac{(\epsilon^{-2}\sin^2\chi + \cos^2\chi) l}{4} \times \frac{\mathrm{erfc}\{(1/\sqrt{2}\,\sigma)\ln[(\epsilon^{-2}\sin^2\chi + \cos^2\chi)^{1/2} l/2a_m]\}}{a_m^{\,2}\exp(2\sigma^2)} \tag{7}$$

from Eq. (4), where $\mathrm{erfc}(x)$ is the complementary error function. The ray distribution becomes

$$p_{\mathrm{pol,rn}}(l) = \frac{\int_0^1 p_{\mathrm{pol}}(l)\pi\epsilon^2(\epsilon^{-2}\sin^2\chi + \cos^2\chi)^{1/2}\mathrm{d}\cos\chi}{\int_0^1 \pi\epsilon^2(\epsilon^{-2}\sin^2\chi + \cos^2\chi)^{1/2}\mathrm{d}\cos\chi} \tag{8}$$

for such particles randomly oriented where the projection area of the cylinder is proportional to $\pi\epsilon^2(\epsilon^{-2}\sin^2\chi + \cos^2\chi)^{1/2}$.[5] It is worth noting here that the ray distribution for a simple spheroid at a fixed orientation [Eq. (5)] is triangular, regardless of the axial ratio of the spheroid. This fundamental geometrical characteristics facilitates a simple rescaling of the radius to calculate the optical efficiencies from a sphere for a spheroid.[5]

The ray distributions from a single spheroid, a single randomly oriented spheroid, a system of polydisperse spheroids at a fixed orientation, and a system of randomly oriented polydisperse spheroids are plotted in Fig. 1. It is clear from the figure that the shape characteristics of an individual particle are washed out by the averaging over the polydispersity and the orientation of the particle. The shape characteristics of an individual particle are expected to be further washed out if particles of different shapes are involved. Thus the ray distribution $p(l)$ of a system of particles such as a bacterial suspension, biological cells, or cirrus clouds where particles are polydisperse, randomly oriented, and (or) of multiple shapes approaches a probability-density function $p(l)$ that is characterized essentially by the mean geometrical path $\langle l\rangle = \int l p(l)\mathrm{d}l$ and the mean-squared geometrical path $\langle l^2\rangle = \int l^2 p(l)\,\mathrm{d}l$ of rays inside the particles. One natural choice of $p(l)$ here is the Gaussian probability-distribution function, which follows the same spirit as the well-known central-limit theorem.[8] We should point out that this choice does not satisfy $p(l < 0) = 0$, but the contribution from near the $l = 0$ region in the ray distribution is much smaller than that from other regions and hence can be ignored.

Let us now assume that the ray distribution is given by a Gaussian distribution:

$$p(x) = \frac{1}{\sqrt{2\pi}\,\sigma}\exp\left[-\frac{(x-\mu)^2}{2\sigma^2}\right]. \tag{9}$$

The extinction and scattering efficiencies are then given by

$$Q_{\mathrm{ext}} = 2 - 2\cos[k(m_r - 1)(\mu - k\sigma^2 m_i)] \times \exp\left\{-k\mu m_i - \frac{k^2\sigma^2[(m_r-1)^2 - m_i^{\,2}]}{2}\right\},$$
$$Q_{\mathrm{abs}} = 1 - \exp[-2km_i(\mu - km_i\sigma^2)] \tag{10}$$

from Eqs. (3) after a straightforward integration. The optical efficiencies [Eqs. (10)], in the intermediate case limit [$k(m_r - 1)l \ll 1$ and $km_i l \ll 1$, where l is the geometrical path],[9] reduce to

$$Q_{\mathrm{ext}} = 2km_i\langle l\rangle + k^2[(m_r-1)^2 - m_i^{\,2}]\langle l^2\rangle,$$
$$Q_{\mathrm{abs}} = 2km_i\langle l\rangle - 2k^2 m_i^{\,2}\langle l^2\rangle,$$
$$Q_{\mathrm{sca}} = k^2|m-1|^2\langle l^2\rangle, \tag{11}$$

where the mean and the mean-squared geometrical paths are given by $\langle l\rangle = \mu$ and $\langle l^2\rangle = \mu^2 + \sigma^2$,

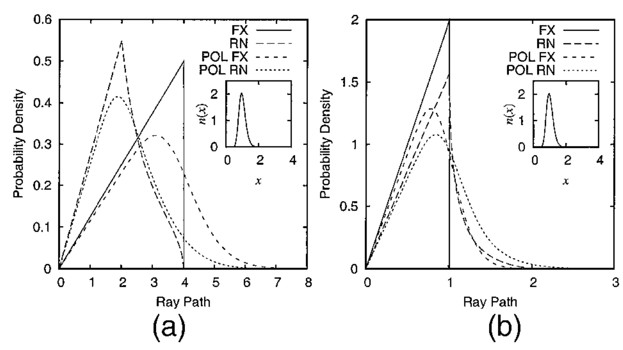

Fig. 1. Ray distributions for a spheroid at a fixed orientation $\chi = 0$ (FX), randomly oriented (RN), polydisperse at a fixed orientation (POL FX), and randomly oriented polydisperse (POL RN). The axial ratio of the spheroid is (a) $\epsilon = 2$ and (b) $\epsilon = 0.5$. Log-normal size distribution $n(x)$ with $a_m = 1$ and $\sigma = 0.2$ is also plotted as insets.

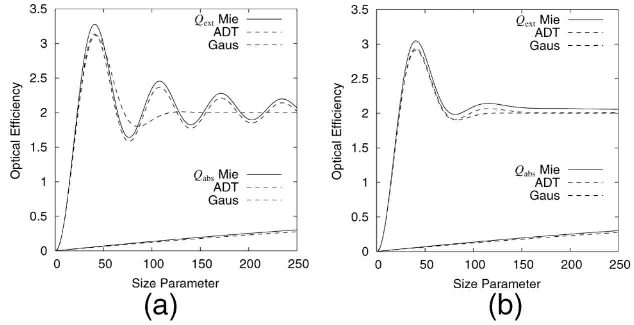

Fig. 2. Extinction and absorption efficiencies of (a) a sphere and (b) a polydisperse sphere with a log-normal radius distribution of $a_m = 1$ and $\sigma = 0.2$ calculated with Mie, ADT, and Gaussian ray approximations. Complex refractive index, $m = 1.05 - i0.0005$. The size distribution has already been shown as insets in Fig. 1.

respectively. These results agree exactly with those for the intermediate region over which the Rayleigh–Gans approximation and the anomalous-diffraction approximation of light scattering from small particles overlap.[1,9] This means that Eqs. (10) from our Gaussian ray approximation reduce to the exact ADT in the intermediate case.

Figure 2 compares the extinction and absorption efficiencies calculated by the exact Mie theory, the exact ADT [Eqs. (1) and (3)], and our Gaussian ray approximation [Eqs. (10)] for a weakly absorbing sphere and a system of the same spheres with a log-normal radius distribution [Eq. (6)] of $a_m = 1$ and $\sigma = 0.2$. Both our Gaussian ray approximation and the ADT, unlike the exact Mie calculation, tend to underestimate the optical efficiencies. This fact is well known.[10,11] The absorption efficiency from our Gaussian ray approximation agrees extremely well with the ADT; at most it differs by 2% from the exact Mie calculation in this comparison. The extinction efficiency agrees well with the exact Mie calculation in the intermediate region for both single spheres and polydisperse spheres, as expected. The Gaussian ray approximation for the polydisperse spheres approaches the exact ADT calculation with maximum relative errors of 3.5% compared to the ADT and of 7% compared to Mie theory.

From our statistical analysis of the anomalous-diffraction theory of light extinction, light extinction depends solely on the probability distribution of the geometrical paths of individual rays inside the particles rather than on the size or the shape of an individual particle. Thus the optical efficiency equivalence[12] can easily be achieved from different-shaped particles or particles of different size distributions as long as they share a common geometrical path distribution of rays.

The geometrical path distribution of rays can be approximated by a Gaussian probability distribution function for a system of particles in which the particles are randomly oriented, polydisperse, and (or) multiple shaped. For such a system of particles the light-extinction measurements essentially determine the mean and the mean-squared geometrical paths of rays from all particles in the system. The shape and size of an individual particle can be deduced only with *a priori* information on the shape and (or) the size distribution of the particles involved. The pursuit of the mean and the mean-squared paths from fitting Eqs. (10) to experimental data, or the general geometrical path distribution of rays $p(l)$ of particles from solving the inverse problem in Eqs. (3), provides an alternative approach to particle sizing and shaping. We note that we have restricted this study to extinction of light from particles of the same type (a common refractive index, m). This statistical interpretation of ADT opens a new way to calculate optical efficiencies of soft particles of different shapes by use of the probability distribution of the geometrical paths of individual rays inside particles.

We thank A. Katz for useful discussions on light scattering from bacterial suspensions. This study is supported in part by the U.S. Department of the Army (grant DAMD17-02-1-0516). The U.S. Army Medical Research Acquisition Activity, 820 Chandler Street, Fort Detrick, Maryland 21702-5014 is the awarding and administering acquisition office. Additional support is given by the U.S. Department of Defense (grants DAMD01-0084 and DAMD98-8147). M. Xu's e-mail address is minxu@sci.ccny.cuny.edu.

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