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Superposition rule for light scattering by a composite particle

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A superposition rule for light scattering by composite particles is presented that expresses the scattering amplitude of a composite particle as a superposition of that of the host particle and those of the shadowed inclusions. The superposition rule is derived for a soft composite particle but also provides insight into light scattering by a general composite scatterer. Favorable comparison with an exact numerical method demonstrates the usefulness of the rule in analyzing light scattering by composite particles such as biological cells.

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Light interaction with small particles is the foundation for remote sensing and biomedical optical spectroscopy and imaging. Mie theory¹ is widely used for spherical uniform particles. For nonspherical particles or composite particles, there is no analytical exact solution available, and numerical methods are commonly required.² Light scattering from composite particles such as aerosol particles³ and cells,⁴ which contain internal structures, is of particular interest in atmospheric science and biomedical applications. For example, the change of the nuclear structure in a cell during neoplastic progression may be probed optically.⁵ Computation of light scattering by a composite particle with inclusions is much more difficult than that for a uniform particle.^{2,4,6,7} The effect of the presence of inclusions on the light scattering property is hard to quantify. In this Letter, a superposition rule for light scattering by composite particles is presented. The superposition rule shows that light scattering by a composite particle can be constructed from a superposition of light scattering by the host particle and those by the shadowed inclusions. This rule applies to composite particles that are optically soft (relative refractive index $|m-1| \ll 1$) when the shadowing of the host particle on inclusions is not far from uniformity. However, it also yields valuable insight into light scattering by a general composite particle.

Light scattering by a soft particle can be well described by the anomalous diffraction theory (ADT) of van de Hulst.^{1,8,9} The applicability of ADT extends to nonsoft particles.^{10,11} The scattering amplitude function in ADT is given by

$$S(\mathbf{q}) = \frac{k^2}{2\pi} \int \{1 - \exp[-ip(\xi, \eta)]\} e^{-i(\xi q_\xi + \eta q_\eta)} d\xi d\eta, \quad (1)$$

where the integration of ξ and η is over the area of the projection of the particle in the direction (z axis) of the incident light, $p(\xi, \eta) = k \int dz [m(\xi, \eta, z) - 1]$ is the phase delay of the ray piercing the particle at the position (ξ, η) , $\mathbf{q} = (q_\xi, q_\eta, 0) = q(\cos \phi, \sin \phi, 0)$ is the wave-vector transfer with a magnitude $2k \sin(\theta/2)$, k

is the wavenumber, and θ , ϕ are the polar and azimuthal angles of scattering, respectively. For a composite particle of the refractive index m_0 containing a region (nucleus) of refractive index $m_1 = m_0 + \delta m$, the phase delay p can be written as the sum of two parts: the phase delay in the host particle $p_0 = k \int dz [m_0(\xi, \eta, z) - 1]$ and the additional phase delay due to the nucleus $\delta p(\xi, \eta) = k \int dz \delta m(\xi, \eta, z)$ [see Fig. 1(a)]. As a consequence, the scattering amplitude function can be written as

$$S(\mathbf{q}) = S_0(\mathbf{q}) + \exp(-i\mathbf{r}_c \cdot \mathbf{q}) f_c S_n(\mathbf{q}), \quad (2)$$

where S_0 and S_n are the scattering amplitude functions for the host particle of refractive index m_0 and the shadowed nucleus of refractive index $\delta m + 1$, respectively, $\mathbf{r}_c = (\xi_c, \eta_c, z_c)$ is the position of the center of the nucleus relative to the center of the host particle, f_c is an operator, and $f_c S_n(\theta)$ is defined by

$$f_c S_n(\mathbf{q}) = \frac{k^2}{2\pi} \int f_c(\xi', \eta') \{1 - \exp[-i\delta p(\xi', \eta')]\} \times e^{-i(\xi' q_\xi + \eta' q_\eta)} d\xi' d\eta', \quad (3)$$

with $f_c(\xi', \eta') = \exp[-ip_0(\xi' + \xi_c, \eta' + \eta_c)]$.

The essence of ADT is that light penetrates straight through the scatterer with only a minimal bending of the ray. Hence light scattering by a composite particle is equivalent to that by two individual entities (the host particle and the nucleus), where the nucleus outside is shadowed by the host particle [see Fig. 1(b)]. If the maximum difference $(\Delta\rho)_{\max}$ in the phase delay $p_0(\xi' + \xi_c, \eta' + \eta_c)$ of the illuminating light on the nucleus due to the host particle is small, then we can replace f_c in Eq. (3) by a constant $\exp(-i\bar{p})$, where \bar{p} is the average phase delay caused by the host particle. If $(\Delta\rho)_{\max} \gg 1$, different regions of the shadowed nucleus are illuminated by light in phase and out of phase alternatively. Since light scattering from volumes of the nucleus illuminated by light out of phase tends to cancel each other, light scattering by the shadowed nucleus is then much suppressed and quite different from that found when it is stand-alone.

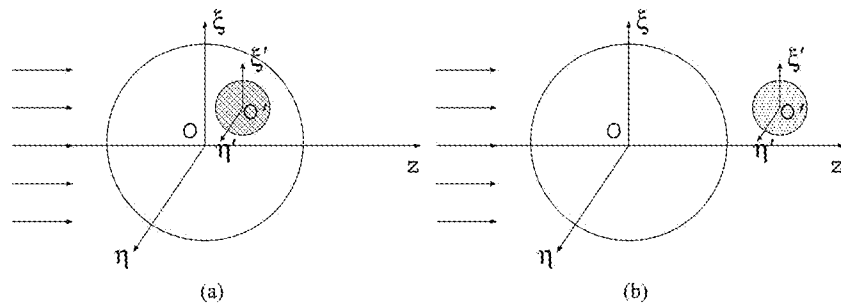


Fig. 1. (a) Composite scatterer illuminated by an incident beam along the z axis. (b) Equivalent configuration with an outside nucleus shadowed by the host particle. The shadowed nucleus has a modified refractive index.

Based on the above observation, the superposition rule asserts that the light scattering amplitude for a composite particle can be expressed as a superposition of those of the host particle and the shadowed nucleus:

$$S(\mathbf{q}) = S_0(\mathbf{q}) + \exp(-i\mathbf{r}_c \cdot \mathbf{q} - i\bar{\rho})S_n(\mathbf{q}), \quad (4)$$

where $\bar{\rho}$ is the average phase delay seen by the nucleus due to the host particle as long as $(\Delta\rho)_{\max} < 1$. The shadowed nucleus is the same as the real nucleus except that its relative refractive index is replaced by $m_1 - m_0 + 1$. If the host particle contains multiple inclusions, Eq. (4) becomes

$$S(\mathbf{q}) = S_0(\mathbf{q}) + \sum_j \exp(-i\mathbf{r}_{c_j} \cdot \mathbf{q} - i\bar{\rho}_j)S_j(\mathbf{q}), \quad (5)$$

where $\bar{\rho}_j$ is the average phase delay seen by the j th inclusion centered at \mathbf{r}_{c_j} due to the host particle (and possibly other inclusions), and S_j is the scattering amplitude function for the j th shadowed inclusion (with a modified refractive index) provided $(\Delta\rho)_{\max} < 1$ is satisfied by each inclusion. The superposition rule extends to the scattering amplitude matrix¹ in which S in Eqs. (4) and (5) is a 2×2 matrix. After obtaining the scattering amplitude of a composite particle, its scattering properties such as optical efficiencies, phase function, and Muller matrix will be fully determined.¹

The application of this rule for a concentric sphere will be demonstrated. Denote the radius of the host particle a and the radius of the nucleus b . The average phase delay is $\bar{\rho} = \frac{2}{3}\rho_{\max}\gamma^{-2}[1 - (1 - \gamma^2)^{3/2}]$, and the maximum phase delay difference is $(\Delta\rho)_{\max} = \frac{1}{2}\rho_{\max}\gamma^2$, where $\rho_{\max} = 2k(m_0 - 1)a$ and $\gamma = b/a$. The superposition rule can be applied when $\frac{1}{2}\rho_{\max}\gamma^2 < 1$.

Figure 2 displays the light extinction efficiencies for various concentric spheres computed exactly¹² and approximately by using the superposition rule, where the scattering amplitudes S_0 and S_n in Eq. (4) are computed by using the MIEV code.¹³ Light extinction spectra for two concentric spheres of size $a = 8 \mu\text{m}$ [$(\Delta\rho)_{\max} = 0.7$], and $a = 32 \mu\text{m}$ [$(\Delta\rho)_{\max} = 2.7$] are shown for the wavelength 500–1000 nm in vacuum. Even when $(\Delta\rho)_{\max} \sim \pi$, the superposition rule still gives an acceptable approximation to light extinction by a composite scatterer.

Figure 3 displays the intensity of light scattering into different directions (unnormalized phase func-

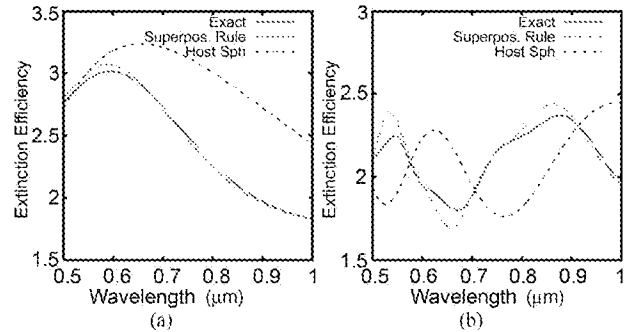


Fig. 2. (Color online) Extinction efficiencies for various concentric spheres of radius (a) $a = 8 \mu\text{m}$ and (b) $a = 32 \mu\text{m}$. The ratio of the size of the nucleus over that of the host particle is $\gamma = 0.5$. The relative refractive indices are $m_0 = 1.02$ for the host particle and $m_1 = 1.08$ for the nucleus. The refractive index of the environment is assumed to be 1.33. The values obtained by using the superposition rule agree well with the exact ones. The extinction efficiencies for the host particle alone are also plotted for comparison.

tion) computed exactly and approximately by using the superposition rule for various concentric spheres. The agreement between the spectra obtained by the superposition rule and the exact ones is excellent.

Light scattering by a composite scatterer with an eccentric inclusion may depend on both the polar and azimuthal angles of scattering. The superposition rule reproduces the phenomenon⁷ that a lower refractive index inclusion causes a secondary diffraction ring to appear on its opposite side. The superposition rule can also be used to compute light scattering by a composite particle with a randomly placed inclusion in a straightforward fashion. One example of practical importance is a spherical scatterer with a nucleus randomly placed inside emulating a biological cell. The scattering amplitude function of the composite scatterer can be expressed as

$$S(\mathbf{q}) = S_0(\mathbf{q}) + f(\mathbf{q}, \mathbf{r}_c)S_n(\mathbf{q}), \quad (6)$$

where $f(\mathbf{q}, \mathbf{r}_c) = \exp[-i\mathbf{r}_c \cdot \mathbf{q} - i\rho_{\max}\sqrt{1 - (\xi_c^2 + \eta_c^2)/a^2}]$ and $\rho_{\max} = 2k(m_0 - 1)a$ when the nucleus is located at \mathbf{r}_c . The average phase delay $\bar{\rho}$ has been replaced by the phase delay for the ray passing through the center of the nucleus for simplicity. The scattering amplitude $\bar{S}(\mathbf{q})$ of the composite sphere with a randomly placed inclusion is obtained by performing a configurational average of $f(\mathbf{q}, \mathbf{r}_c)$ in Eq. (6) over \mathbf{r}_c , which yields

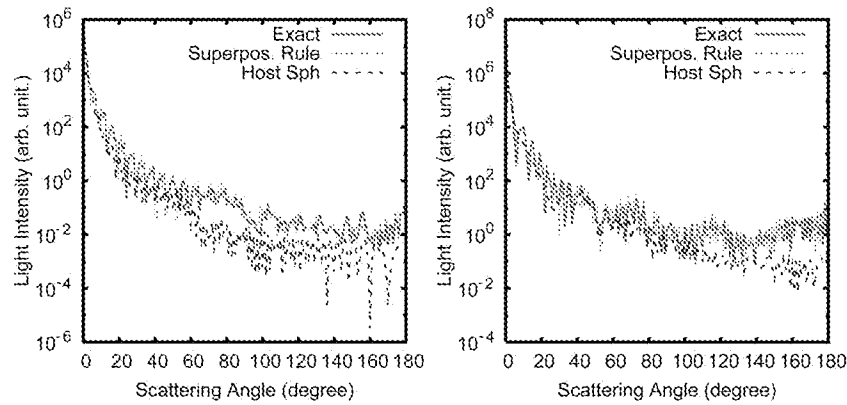


Fig. 3. (Color online) Angular light scattering by various concentric spheres of the same parameters as those in Fig. 2. The wavelength of the incident light is 500 nm. The angular spectra obtained by using the superposition rule agree well with the exact ones. The angular spectra for the host particle alone are also plotted for comparison.

$$\overline{f(\mathbf{q})} = 3 \int_0^{\pi/2} \exp(-i\rho_{\max}\sqrt{1-\epsilon^2\cos^2\tau}) \times J_0(qa\epsilon\cos\tau)\cos\tau\sin^2\pi d\tau, \quad (7)$$

dependent on ϵa , the maximum displacement of the nucleus from the center of the host. The behavior of $\overline{S(\mathbf{q})}$ departs from that of a concentric sphere when $\epsilon > 0$. It is noted that the phase function of such a composite particle is determined by $|\overline{S(\mathbf{q})}|^2 = |S_0(\mathbf{q})|^2 + |S_n(\mathbf{q})|^2 + 2\Re[\overline{f(\mathbf{q})}S_0^*(\mathbf{q})S_n(\mathbf{q})] \neq |\overline{S(\mathbf{q})}|^2$, where \Re and $*$ denote the real part and the complex conjugate, respectively.

The superposition rule provides a simple means to analyze light scattering by a soft complex particle and to quantify the contributions from each individual inclusion. It is, in particular, suited to investigate light scattering by a biological cell and its internal structure (nucleus, mitochondria, and other organelles). Since the scattering amplitude fully determines the (vector) light scattering properties by a scatterer, it is believed the superposition rule about the scattering amplitude will be important to understand light scattering by a composite particle and facilitate noninvasive probing of such particles using light.

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