Angle- and size-dependent characteristics of incoherent Raman and fluorescent scattering by microspheres. 1. General expressions

Vadim Griaznov, Igor Veselovskii, Alexei Kolgotin, and David N. Whiteman

A dipole model is used to simulate incoherent Raman and fluorescent scattering by microspheres. The use of the addition theorem for spherical harmonics circumvents the need to evaluate double sums in the final formulas, thereby drastically reducing computational effort. Special attention is paid to consideration of backscattering geometry, which is important for lidar applications. The formulas derived for backscattering geometry decrease the computation time for size parameter $x \sim 100$ by a factor of 200 compared with the time for calculations performed at other angles. © 2002 Optical Society of America

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1. Introduction

The inelastic scattering of laser radiation from microspheres is a powerful technique for gaining information about particle size and chemical composition.[1,2] Nevertheless, quantitative evaluation of inelastic scattering spectra entails serious complications because the radiation field within the microparticle depends in a complicated way on particle size, refractive index, and wavelength. To relate the measured inelastic spectrum to the physical parameters of the particle, one should use the appropriate physical model. Such a model, based on embedded dipoles, was suggested and investigated by Chew *et al.*,[3,4] Kerker *et al.*,[5,6] Chew,[7] Kerker and Druger,[8] and Druger and McNulty.[9] In a dipole model the electric field transmitted in the particle induces classic electric dipoles in molecules. These dipoles emit frequency-shifted inelastically scattered radiation. Owing to the incoherence of the scattering process, the total power emitted to the far field by the whole set of reradiating molecules is obtained by integration of all dipole powers. But the numerical integration of the expressions derived by Kerker *et al.*[6] is extremely time consuming, so these earlier results (obtained 23 years ago, when computer technology was much less advanced) were limited to relatively small size parameters of $x \sim 20$.

To decrease computation time, simplifications of the dipole model were suggested. For a rotationally symmetric distribution of molecules the volume integration can be split into solid-angle integration and integration over the radial position. It has been shown that solid-angle integration can be solved analytically.[7] Thus the formula for the inelastically scattered power is reduced to an integral of one dimension, which drastically reduces the computational effort compared with that required for numerical integration over a three-dimensional distribution of molecules.[10] Still, the consideration of integrated inelastic scattering does not permit the study of the scattering phase function and the polarization characteristics of the scattered radiation.

One may achieve some simplifications and reduction of computation time by considering rapidly rotating molecules,[9,11] thus escaping the need to address vector relations between the excitation field and molecular emission. Such averaging over dipole orientations should lead to correct results for fluorescence in many cases, but in Raman scattering the dipoles are field oriented rather than randomly distributed, so at least a comparison between the results obtained with these approaches should be made.

In this paper we derive the formulas for calculation of incoherent Raman scattering by microspheres, using the classic dipole model. The application of the addition theorem circumvents the need for double summing in the final formulas for inelastic scattering,

V. Griaznov, I. Veselovskii (katyv@orc.ru), and A. Kolgotin are with the Physics Instrumentation Center of the General Physics Institute, Troitsk, Moscow Region, 142190, Russia. D. N. Whiteman is with the NASA Goddard Space Flight Center, Greenbelt, Maryland 20771-0001.

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thus reducing the computation time. With these formulas it is possible to calculate the scattering phase functions and intensity of morphology-dependent resonances for different polarizations of incident radiation and for a wide range of size parameters.

2. Model

The calculations were carried out with the assumptions that the induced dipoles are parallel to the exciting field and that an active molecule does not rotate during the interval between excitation and emission. This approach applies most clearly to fluorescence when orientations of dipoles are fixed and to Raman scattering when molecules are isotropically polarizable. The active molecules are assumed to be uniformly distributed through the microsphere. Although this model corresponds to a rather idealized system, it allows us to illustrate the major effects that depend on the morphology and optical properties of particles.

A. General Expressions

A plane wave of frequency ω_0 propagating along the z axis in a medium with refractive index n_2 may be expanded in terms of vector spherical harmonics[12–14]:

$$\mathbf{E}_{\text{inc}}(\mathbf{r}) = \sum_{l'm'} \Bigg\{ \frac{ic}{n_2{}^2\omega_0} \alpha_E(l', m')\nabla \times [j_{l'}(k_{02}r)\mathbf{X}_{l'm'}(\theta, \varphi)] + \alpha_M(l', m')j_{l'}(k_{02}r)\mathbf{X}_{l'm'}(\theta, \varphi) \Bigg\}, \tag{1}$$

$$\mathbf{B}_{\text{inc}}(\mathbf{r}) = \sum_{l'm'} \Bigg\{ \alpha_E(l', m')\nabla \times [j_{l'}(k_{02}r)\mathbf{X}_{l'm'}(\theta, \varphi)] - \frac{ic}{\omega_0} \alpha_M(l', m')j_{l'}(k_{02}r)\mathbf{X}_{l'm'}(\theta, \varphi) \Bigg\}. \tag{2}$$

For horizontal $\mathbf{E}_{\text{inc}}{}^{OX}$ and vertical $\mathbf{E}_{\text{inc}}{}^{OY}$ polarization of the incident radiation with respect to the scattering plane, taken as the x–z plane, we have

$$\mathbf{E}_{\text{inc}}{}^{OX} = E_0\mathbf{e}_x \exp(ik_{02}z),$$
$$\mathbf{E}_{\text{inc}}{}^{OY} = E_0\mathbf{e}_y \exp(ik_{02}z).$$

The expansion coefficients are

$$\alpha_E{}^{OX}(l', m') = i^{l'}[4\pi(2l' + 1)]^{1/2} \frac{\delta_{m',l} - \delta_{m',-1}}{2i} n_2E_0,$$

$$\alpha_M{}^{OX}(l', m') = i^{l'}[4\pi(2l' + 1)]^{1/2} \frac{\delta_{m',1} + \delta_{m',-1}}{2} E_0,$$

$$\alpha_E{}^{OY}(l', m') = -i^{l'}[4\pi(2l' + 1)]^{1/2} \times \frac{\delta_{m',1} + \delta_{m',-1}}{2} n_2E_0,$$

$$\alpha_M{}^{OY}(l', m') = i^{l'}[4\pi(2l' + 1)]^{1/2} \frac{\delta_{m',1} - \delta_{m',-1}}{2i} E_0.$$

Here $k_{02} = n_2\omega_0/c$ is the wave number of the outer medium and $j_{l'}(k_{02}r)$ denote spherical Bessel functions. All the expressions here and below are written in the Gaussian system of units; the time-dependent factor is $\exp(-i\omega t)$.

The field transmitted inside the sphere of radius a and optical properties n_1 and μ_1 located at the origin of the coordinate system is expanded as[13,14]

$$\mathbf{E}_{\text{tra}}(\mathbf{r}') = \sum_{l'm'} \Bigg\{ \frac{ic}{n_1{}^2\omega_0} \gamma_E(l', m')\nabla \times [j_{l'}(k_{01}r')\mathbf{X}_{l'm'}(\theta', \varphi')] + \gamma_M(l', m')j_{l'}(k_{01}r')\mathbf{X}_{l'm'}(\theta', \varphi') \Bigg\}, \tag{3}$$

with coefficients

$$\gamma_E(l', m') = g_E(l')\alpha_E(l', m'),$$
$$g_E(l') = \frac{i\mu_1M^2}{\mu_2M\psi_{l'}(Mx)\xi_{l'}{}'(x) - \mu_1\xi_{l'}(x)\psi_{l'}{}'(Mx)},$$
$$\gamma_M(l', m') = g_M(l')\alpha_M(l', m')$$
$$g_M(l') = \frac{i\mu_1M}{\mu_1\psi_{l'}(Mx)\xi_{l'}{}'(x) - \mu_2M\xi_{l'}(x)\psi_{l'}{}'(Mx)},$$

where $\psi_l(\rho) = \rho j_l(\rho)$, $\xi_l(\rho) = \rho h_l{}^{(1)}(\rho)$, $\psi_l{}'(\rho)$, $\xi_l{}'(\rho)$ are Riccati–Bessel functions and their derivatives, $h_l{}^{(1)}(\rho)$ denotes Hankel functions of the first kind; $k_{01} = (\omega_0/c)n_1$ is the wave number inside a particle, $M = n_1/n_2 = k_{01}/k_{02}$ is the relative refractive index, and $x = k_{02}a$. We use mainly Riccati–Bessel functions instead of spherical Bessel functions because the former are preferable in programming and numerical calculations.

Assuming that the polarizibility of medium α_1 is isotropic, the electric dipole moment induced by a transmitted field is defined by $\alpha_1\mathbf{E}_{\text{tra}}(r)$. The radiation emission at frequency ω by the assembly of dipoles is the reason for the presence of inelastically scattered field $\mathbf{E}_2(\omega)$ that arises in the outer medium. Assuming that the relative change in frequency $p = \omega_0/\omega$ is not too large, we can assume that $n_1(\omega) = n_1(\omega_0)$. In the far zone an electric field of radiation scattered inelastically from a spherical particle owing to a single dipole is given by[3,5,6,10]

$$\mathbf{E}_2{}^1 = \frac{\exp(ik_2r)}{k_2r} \sum_{l,m} (-i)^{l+1}\Bigg[c_M{}^1(l, m)\mathbf{X}_{lm}(\theta, \varphi) - \frac{1}{n_2} c_E{}^1(l, m)\mathbf{e}_r \times \mathbf{X}_{lm}(\theta, \varphi) \Bigg]. \tag{4}$$

A superscript in our notation means that the field is produced by a single dipole; the superscript is omitted after the implementation of volume integration. The coefficients $c_E{}^1(l, m)$ and $c_M{}^1(l, m)$ were derived in Ref. 3 and are expressed here in terms of Riccati–Bessel functions:

$$c_E^1(l, m) = d_E(l) a_E^1(l, m),$$

$$d_E(l) = \frac{i\mu_2}{M[\mu_2 M \psi_l(pMx)\xi_l'(px) - \mu_1 \xi_l(px)\psi_l'(pMx)]},$$

$$c_M^1(l, m) = d_M(l) a_M^1(l, m),$$

$$d_M(l) = \frac{i\mu_2}{\mu_1 \psi_l(pMx)\xi_l'(px) - \mu_2 M \xi_l(px)\psi_l'(pMx)}.$$

The expansion coefficients[7,10]

$$a_E^1(l, m) = 4\pi k^2 M n_2 \mu_1 \alpha_1 \mathbf{E}_{\mathrm{tra}}\{\nabla \times [j_l(pk_{01}r')\mathbf{X}_{lm}^*(\theta', \varphi')]\},$$

$$a_M^1(l, m) = 4\pi i k^3 M n_2 \mu_1 \alpha_1 j_l(pk_{01}r')\mathbf{E}_{\mathrm{tra}} \cdot \mathbf{X}_{lm}^*(\theta', \varphi')$$

determine a dipole field $\mathbf{E}_{\mathrm{dip}}^1$, which is created by an excited molecule located at coordinates (r', θ', φ') inside the scattering particle; $k = \omega/c$. Using the definition of vector spherical harmonics,[12] one can express them in spherical coordinates as

$$\mathbf{X}_{lm}(\theta, \varphi) = \frac{i}{[l(l+1)]^{1/2}} \left[\frac{1}{\sin\theta} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial \varphi} \mathbf{e}_\theta - \frac{\partial Y_{lm}(\theta, \varphi)}{\partial \theta} \mathbf{e}_\varphi \right], \quad (5)$$

where $Y_{lm}(\theta, \varphi)$ are scalar spherical harmonics and $\mathbf{e}_\theta$ and $\mathbf{e}_\varphi$ denote unit vectors of a spherical system. Therefore the components of $\mathbf{E}_2^1(\omega)$ can be written as

$$E_{2\theta}^1 = \frac{\exp(ik_2 r)}{k_2 r} \sum_{l,m} (-i)^{l+1} \frac{i}{[l(l+1)]^{1/2}} \times \left[c_M^1(l, m) \frac{1}{\sin\theta} \frac{\partial Y_{lm}}{\partial \varphi} - \frac{c_E^1(l, m)}{n_2} \frac{\partial Y_{lm}}{\partial \theta} \right], \quad (6)$$

$$E_{2\varphi}^1 = -\frac{\exp(ik_2 r)}{k_2 r} \sum_{lm} (-i)^{l+1} \frac{i}{[l(l+1)]^{1/2}} \times \left[\frac{c_E^1(l, m)}{n_2} \frac{1}{\sin\theta} \frac{\partial Y_{lm}}{\partial \varphi} + c_M^1(l, m) \frac{\partial Y_{lm}}{\partial \theta} \right]. \quad (7)$$

Taking into account expressions for $c_M^1(l, m)$ and $c_E^1(l, m)$, we find the components

$$E_{2\theta}^1 = 4\pi k^3 M n_2 \mu_1 \alpha_1 \frac{\exp(ik_2 r)}{k_2 r} \sum_{l,m} \frac{i(-i)^{l+1}}{[l(l+1)]^{1/2}} \times \Bigg(i d_M(l) \frac{1}{\sin\theta} j_l(pk_{01}r') \frac{\partial Y_{lm}(\theta, \varphi)}{\partial \varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{X}_{lm}^*(\theta', \varphi') - \frac{d_E(l)}{kn_2} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial \theta} \mathbf{E}_1(\mathbf{r}') \cdot \{\nabla \times [j_l(pk_{01}r')\mathbf{X}_{lm}^*(\theta', \varphi')]\} \Bigg), \quad (8)$$

$$E_{2\varphi}^1 = -4\pi k^3 M n_2 \mu_1 \alpha_1 \frac{\exp(ik_2 r)}{k_2 r} \sum_{l,m} \frac{i(-i)^{l+1}}{[l(l+1)]^{1/2}} \times \Bigg(\frac{d_E(l)}{kn_2} \frac{1}{\sin\theta} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial \varphi} \mathbf{E}_1(\mathbf{r}') \cdot \{\nabla \times [j_l(pk_{01}r')\mathbf{X}_{lm}^*(\theta', \varphi')]\} + i d_M(l) j_l(pk_{01}r') \frac{\partial Y_{lm}(\theta, \varphi)}{\partial \theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{X}_{lm}^*(\theta', \varphi') \Bigg). \quad (9)$$

The final goal of our analysis is calculation of the angular scattering cross section of a particle. In the case of a single excited molecule the cross section can be expressed as

$$\frac{d\sigma^1}{d\Omega} = \frac{|\mathbf{E}_2^1|^2}{E_0^2} r^2 = \frac{|E_{2\theta}^1|^2 + |E_{2\varphi}^1|^2}{E_0^2} r^2. \quad (10)$$

Incoherent scattering from the whole volume of a particle in cases of parallel and perpendicular polarization of the incident wave is determined correspondingly by the integral values

$$\frac{d\sigma^{OX}}{d\Omega} = \int_v N \frac{d\sigma^{1,OX}}{d\Omega} d^3r' = \int_v N \frac{|E_{2\theta}^{1,OX}|^2}{E_0^2} r^2 d^3r' + \int_v N \frac{|E_{2\varphi}^{1,OX}|^2}{E_0^2} r^2 d^3r' = \frac{d\sigma_{2\theta}^{OX}}{d\Omega} + \frac{d\sigma_{2\varphi}^{OX}}{d\Omega},$$

$$\frac{d\sigma^{OY}}{d\Omega} = \int_v N \frac{d\sigma^{1,OY}}{d\Omega} d^3r' = \int_v N \frac{|E_{2\theta}^{1,OY}|^2}{E_0^2} r^2 d^3r' + \int_v N \frac{|E_{2\varphi}^{1,OY}|^2}{E_0^2} r^2 d^3r' = \frac{d\sigma_{2\theta}^{OY}}{d\Omega} + \frac{d\sigma_{2\varphi}^{OY}}{d\Omega}, \quad (11)$$

where v is the total volume of a particle and N means the density of dipoles radiating at Raman frequency ω. For the reader's further convenience we set up a correspondence between the differential cross sections used here and the scattered intensities H_H, V_H, H_V, and V_V introduced by Kerker and Druger[8] because this notation is commonly used in simulations and in experimental data analysis:

$$\frac{d\sigma_{2\theta}^{OX}}{d\Omega} \equiv \frac{H_H}{I_{\mathrm{inc}}}, \qquad \frac{d\sigma_{2\varphi}^{OX}}{d\Omega} \equiv \frac{V_H}{I_{\mathrm{inc}}},$$

$$\frac{d\sigma_{2\theta}^{OY}}{d\Omega} \equiv \frac{H_v}{I_{\mathrm{inc}}}, \qquad \frac{d\sigma_{2\varphi}^{OY}}{d\Omega} \equiv \frac{V_V}{I_{\mathrm{inc}}},$$

where I_{inc} is the intensity of the incident radiation.

Unfortunately, the number of multipole coefficients taken into consideration is proportional to $(lm)_{\max} = l_{\max}^2$, where $l_{\max}$ is the maximum value of l, $l_{\max} = x + 4x^{1/3} + 2$.[14] The number of multipole terms can be reduced essentially if we employ the addition theorem[12] and escape from double sums in the expression for $\mathbf{E}_2{}^1(\mathbf{r})$. The details of this transition are presented in Appendix A. Further, the expressions for $E_{2\theta}{}^1$ and $E_{2\varphi}{}^1$ can be simplified additionally if we apply a similar representation of vector spherical harmonics expressed in spherical coordinates for transmitted field $\mathbf{E}_{\text{tra}}(\mathbf{r}')$. For OX and OY polarization of the incident wave the transmitted field may be written as

$$\mathbf{E}_{\text{tra}}{}^{OX}(\mathbf{r}') = E_0[\mathbf{e}_r A_r(\theta', \delta')\cos\varphi' + \mathbf{e}_\theta A_\theta(\theta', \delta')\cos\varphi' + \mathbf{e}_\varphi A_\varphi(\theta', \delta')\sin\varphi'], \tag{12}$$

$$\mathbf{E}_{\text{tra}}{}^{OY}(\mathbf{r}') = E_0[\mathbf{e}_r A_r(\theta', \delta')\sin\varphi' + \mathbf{e}_\theta A_\theta(\theta', \delta')\sin\varphi' - \mathbf{e}_\varphi A_\varphi(\theta', \delta')\cos\varphi'], \tag{13}$$

where

$$A_r(\theta', \delta') = \sum_{l'=1}^{\infty} i^{l'}(2l'+1)\frac{i}{M} g_E(l')\frac{\psi_{l'}(Mx\delta')}{(Mx\delta')^2} \times \rho_{l'}{}^1(\theta')\sin\theta', \tag{14}$$

$$A_\theta(\theta', \delta') = \sum_{l'=1}^{\infty} i^{l'}\frac{(2l'+1)}{l'(l'+1)} \times \left[\frac{i}{M} g_E(l')\psi_{l'}{}'(Mx\delta')\tau_{l'}{}^1(\theta') - g_M(l')\psi_{l'}(Mx\delta')\rho_{l'}{}^1(\theta')\right]\frac{1}{Mx\delta'}, \tag{15}$$

$$A_\varphi(\theta', \delta') = \sum_{l'=1}^{\infty} i^{l'}\frac{(2l'+1)}{l'(l'+1)} \times \left[g_M(l')\psi_{l'}(Mx\delta')\tau_{l'}{}^1(\theta') - \frac{i}{M} g_E(l')\psi_{l'}{}'(Mx\delta')\rho_{l'}{}^1(\theta')\right]\frac{1}{Mx\delta'}, \tag{16}$$

where

$$\rho_{l'}{}^1(\theta') = \frac{P_{l'}{}^1(\theta')}{\sin\theta'}, \quad \tau_{l'}{}^1(\theta') = \frac{\mathrm{d}P_{l'}{}^1(\theta')}{\mathrm{d}\theta'}, \quad \delta' = \frac{r'}{a},$$

and $P_{l'}{}^1(\theta')$ is the associated Legendre function.

Taking into account the results of Appendix A, we have finally

$$E_{2\theta}{}^{1OX} = E_0\frac{\exp(ik_2r)}{k_2r}k^3Mn_2\mu_1\alpha_1 \times \sum_{l=1}^{\infty}(-i)^{l+1}\frac{2l+1}{l(l+1)}\Bigg[iA_\theta(\theta')d_M(l) \times \frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\varphi\varphi}\cos\varphi' - iA_\varphi(\theta')d_M(l)\frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\theta\varphi}\sin\varphi' - l(l+1)MA_r(\theta')d_E(l)\frac{\psi_l(pMx\delta')}{(pMx\delta')^2}S_\theta\cos\varphi' - MA_\varphi(\theta')d_E(l)\frac{\psi_l{}'(pMx\delta')}{pMx\delta'} \times S_{\varphi\theta}\sin\varphi' - MA_\theta(\theta')d_E(l) \times \frac{\psi_l{}'(pMx\delta')}{pMx\delta'}S_{\theta\theta}\cos\varphi'\Bigg], \tag{17}$$

$$E_{2\varphi}{}^{1OX} = E_0\frac{\exp(ik_2r)}{k_2r}k^3Mn_2\mu_1\alpha_1\sum_{l=1}^{\infty} \times (-i)^{l+1}\frac{2l+1}{l(l+1)}\Bigg[-iA_\theta(\theta')d_M(l) \times \frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\varphi\theta}\cos\varphi' + iA_\varphi(\theta')d_M(l)\frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\theta\theta}\sin\varphi' - l(l+1)MA_r(\theta')d_E(l)\frac{\psi_l(pMx\delta')}{(pMx\delta')^2}S_\varphi\cos\varphi' - MA_\varphi(\theta')d_E(l)\frac{\psi_l{}'(pMx\delta')}{pMx\delta'}S_{\varphi\varphi}\sin\varphi' - MA_\theta(\theta')d_E(l)\frac{\psi_l{}'(pMx\delta')}{pMx\delta'}S_{\theta\varphi}\cos\varphi'\Bigg], \tag{18}$$

$$E_{2\theta}{}^{1OY} = E_0\frac{\exp(ik_2r)}{k_2r}k^3Mn_2\mu_1\alpha_1 \times \sum_{l=1}^{\infty}(-i)^{l+1}\frac{2l+1}{l(l+1)}\Bigg[iA_\theta(\theta')d_M(l) \times \frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\varphi\varphi}\sin\varphi' + iA_\varphi(\theta')d_M(l)\frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\theta\varphi}\cos\varphi' - l(l+1)MA_r(\theta')d_E(l)\frac{\psi_l(pMx\delta')}{(pMx\delta')^2}S_\theta\sin\varphi' + MA_\varphi(\theta')d_E(l)\frac{\psi_l{}'(pMx\delta')}{pMx\delta'}S_{\varphi\theta}\cos\varphi' - MA_\theta(\theta')d_E(l)\frac{\psi_l{}'(pMx\delta')}{pMx\delta'}S_{\theta\theta}\sin\varphi'\Bigg], \tag{19}$$

$$E_{2\varphi}{}^{1OY} = E_0\frac{\exp(ik_2r)}{k_2r}k^3Mn_2\mu_1\alpha_1\sum_{l=1}^{\infty} \times (-i)^{l+1}\frac{2l+1}{l(l+1)}\Bigg[-iA_\theta(\theta')d_M(l) \times \frac{\psi_l(pMx\delta')}{pMx\delta'}S_{\varphi\theta}\sin\varphi'$$

$$- iA_{\varphi}(\theta')d_M(l)\,\frac{\psi_l(pMx\delta')}{pMx\delta'}\,S_{\theta\theta}\cos\varphi' - l(l + 1)MA_r(\theta')d_E(l)\,\frac{\psi_l(pMx\delta')}{(pMx\delta')^2}\,S_{\varphi}\sin\varphi' + MA_{\varphi}(\theta')d_E(l)\,\frac{\psi_l'(pMx\delta')}{pMx\delta'}\,S_{\varphi\varphi}\cos\varphi' - MA_{\theta}(\theta')d_E(l)\,\frac{\psi_l'(pMx\delta')}{pMx\delta'}\,S_{\theta\varphi}\sin\varphi'\Bigg]. \quad (20)$$

Depolarization of the scattered radiation for both cases of incident wave polarization can be determined as ratios:

$$\delta^{OX}(\theta, x) = \frac{V_H(\theta, x)}{H_H(\theta, x)}, \quad (21)$$

$$\delta^{OY}(\theta, x) = \frac{H_V(\theta, x)}{V_V(\theta, x)}, \quad (22)$$

To compare the scattering properties of aerosol and bulk media we define the normalized scattering cross section as

$$\frac{\left(\frac{d\sigma}{d\Omega}\right)_{\text{aerosol}}}{\left(\frac{d\sigma}{d\Omega}\right)_{\text{bulk}}}, \quad (23)$$

where in the denominator we imply an inelastic incoherent cross section in the backward direction:

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{bulk}} = \frac{4}{3}\,\pi a^3 N \alpha_1^{\,2} k^4 \mu_1 \mu_2 M. \quad (24)$$

This formula for the differential cross section of a bulk medium may be derived in a manner similar to that described in Refs. 6 and 10. It should be mentioned that, after integration over all directions, we arrive to the same expression as Chew[7] does.

B. Inelastic Scattering by Small Particles

With small radii the expressions for the scattered field can be obtained analytically. For Riccati–Bessel functions of the argument $\rho \to 0$ we can find that[15]

$$\frac{\psi_{l'}(\rho)}{\rho^2} = \begin{cases} 1/3 & l' = 1 \\ 0 & l' > 1 \end{cases},$$

$$\frac{\psi_{l'}'(\rho)}{\rho} = \begin{cases} 2/3 & l' = 1 \\ 0 & l' > 1 \end{cases},$$

$$\frac{\psi_{l'}(\rho)}{\rho} = 0 \qquad \text{any } l'.$$

Taking into account that $\rho_1^{\,1}(\theta') = -1$ and $\tau_1^{\,1}(\theta') = -\cos\theta'$, we obtain for the internal field $\mathbf{E}_1(\mathbf{r}')$

$$A_r = \frac{3}{M^2 + 2}\sin\theta',$$

$$A_{\theta} = \frac{3}{M^2 + 2}\cos\theta',$$

$$A_{\varphi} = -\frac{3}{M^2 + 2}.$$

After some manipulations the components of scattered field are

$$E_{2\theta}^{\;1OX} = \frac{9k^3\mu_1\alpha_1 E_0\exp(ik_2 r)}{(M^2 + 2)^2 k_2 r}\cos\theta,$$

$$E_{2\varphi}^{\;1OX} = 0,$$

$$E_{2\theta}^{\;1OY} = 0,$$

$$E_{2\varphi}^{\;1OY} = \frac{9k^3\mu_1\alpha_1 E_0\exp(ik_2 r)}{(M^2 + 2)^2 k_2 r}.$$

So the depolarization vanishes in the case of small particles. The cross sections are

$$\frac{d\sigma_{\theta}^{\;OX}}{d\Omega} = \frac{216\pi^2 N\alpha_1^{\,2} p^4 x^3 \mu_1^{\,2}}{n_2^{\,6}(M^2 + 2)^4 \lambda_0}\cos^2\theta,$$

$$\frac{d\sigma_{\varphi}^{\;OY}}{d\Omega} = \frac{216\pi^2 N\alpha_1^{\,2} p^4 x^3 \mu_1^{\,2}}{n_2^{\,6}(M^2 + 2)^4 \lambda_0}, \quad (25)$$

where λ_0 is the wavelength of incident radiation. It is obvious from the formulas [Eq. (25)] that the scattering is proportional to volume of particles.

C. Inelastic Scattering in the Backward Direction

The case of special interest is the application of the dipole model to lidar sounding. In the lidar technique, only backscattered radiation is considered, and for $\theta = 180°$ the expressions for the scattered field can be significantly simplified. From the properties of associated Legendre functions we find that[15]

$$\frac{m}{\sin\theta}\,P_l^{\,m}(\theta)\bigg|_{180°} = \frac{dP_l^{\,m}(\theta)}{d\theta}\bigg|_{180°} = 0 \qquad |m| \neq 1, \quad (26)$$

$$\frac{m}{\sin\theta}\,P_l^{\,m}(\theta)\bigg|_{180°} = \frac{dP_l^{\,m}(\theta)}{d\theta}\bigg|_{180°} = (-1)^l\,\frac{1}{2} \quad m = -1, \quad (27)$$

$$\frac{m}{\sin\theta}\,P_l^{\,m}(\theta)\bigg|_{180°} = (-1)^l\,\frac{l(l + 1)}{2},$$

$$\frac{dP_l^{\,m}(\theta)}{d\theta}\bigg|_{180°} = -(-1)^l\,\frac{l(l + 1)}{2} \qquad m = 1. \quad (28)$$

Therefore for backward scattering we do not need to use the addition theorem to get single sums because all the terms with $|m| \neq 1$ are equal to zero. As is shown in Appendix B, the angle-scattering cross section may be analytically integrated over angle φ to yield the expressions

$$\int_0^{2\pi} d\varphi' \frac{d\sigma_\theta^{1OX}}{d\Omega} = \frac{\pi}{4} |k^2 M \mu_1 \alpha_1|^2 \{3[|B_1|^2 + |B_3|^2 + |B_4|^2 - 2\,\mathrm{Im}(B_1 B_3^*) - 2\,\mathrm{Im}(B_1 B_4^*) + 2\,\mathrm{Re}(B_3 B_4^*)] + 3[|B_2|^2 + |B_5|^2 - 2\,\mathrm{Im}(B_2 B_5^*)] + [-2\,\mathrm{Re}(B_1 B_2^*) + 2\,\mathrm{Im}(B_1 B_5^*) + 2\,\mathrm{Im}(B_2 B_3^*) + 2\,\mathrm{Im}(B_2 B_4^*) - 2\,\mathrm{Re}(B_4 B_5^*) - 2\,\mathrm{Re}(B_3 B_5^*)]\}, \tag{29}$$

$$\int_0^{2\pi} d\varphi' \frac{d\sigma_\varphi^{1OX}}{d\Omega} = \frac{\pi}{4} |k^2 M \mu_1 \alpha_1|^2 [|B_1|^2 + |B_2|^2 + |B_3|^2 + |B_4|^2 + |B_5|^2 + 2\,\mathrm{Re}(B_1 B_2^*) - 2\,\mathrm{Im}(B_1 B_3^*) - 2\,\mathrm{Im}(B_1 B_4^*) - 2\,\mathrm{Im}(B_1 B_5^*) - 2\,\mathrm{Im}(B_2 B_3^*) - 2\,\mathrm{Im}(B_2 B_4^*) - 2\,\mathrm{Im}(B_2 B_5^*) + 2\,\mathrm{Re}(B_3 B_4^*) + 2\,\mathrm{Re}(B_3 B_5^*) + 2\,\mathrm{Re}(B_4 B_5^*)], \tag{30}$$

where

$$B_1(\delta', \theta') = A_\theta(\delta', \theta') \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} d_M(l) \times \frac{\psi_l(pMx\delta')}{pMx\delta'} \rho_l^1(\theta'),$$

$$B_2(\delta', \theta') = A_\varphi(\delta', \theta') \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} d_M(l) \times \frac{\psi_l(pMx\delta')}{pMx\delta'} \tau_l^1(\theta'),$$

$$B_3(\delta', \theta') = MA_r(\delta', \theta') \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} l(l+1) d_E(l) \frac{\psi_l(pMx\delta')}{(pMx\delta')^2} \rho_l^1(\theta') \sin\theta',$$

$$B_4(\delta', \theta') = MA_\theta(\delta', \theta') \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} d_E(l) \times \frac{\psi_l'(pMx\delta')}{pMx\delta'} \tau_l^1(\theta'),$$

$$B_5(\delta', \theta') = MA_\varphi(\delta', \theta') \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} d_E(l) \times \frac{\psi_l'(pMx\delta')}{pMx\delta'} \rho_l^1(\theta'). \tag{31}$$

The integration over the rest of the coordinates can be achieved in numerical form. Here we present the expressions for only one polarization of an incident wave because for backscattering the results obtained for both polarizations coincide. The application of expressions (29) and (30) for backscattering geometry decreases the computation time by a factor of 200 compared with that for calculations performed with formulas (17)–(20).

3. Summary

The application of classic dipole model[3–7] formulas to incoherent Raman scattering by microspheres is highly time consuming. Using the addition theorem for spherical harmonics may accelerate the computations, which will circumvent the need to evaluate double sums in final formulas, thereby drastically reducing the computational effort. For backscattering geometry the integration over one of the angles may be performed analytically. For $x \sim 100$ the derived formulas lead to the decrease of computation time by a factor of 200 compared with that required for other angles. The formulas derived make possible the simulation of scattering by particles of much larger sizes than was previously possible. We demonstrate these computational improvements in a further paper.[16]

Appendix A: Application of an Addition Theorem to Expressions for an Inelastically Scattered Field

In pursuing the task of escaping double sums in expressions for $\mathbf{E}_2^{\,1}(\mathbf{r})$, first we write complex conjugate vectors $\mathbf{X}_{lm}^*(\theta', \varphi')$ and $\{\nabla \times [j_l(pK_1 r')\mathbf{X}_{lm}(\theta', \varphi')]\}^*$ through their components in a spherical system:

$$\mathbf{X}_{lm}^*(\theta, \varphi) = \frac{-i}{[l(l+1)]^{1/2}} \times \left[\frac{1}{\sin\theta} \frac{\partial Y_{lm}^*(\theta, \varphi)}{\partial \varphi} \mathbf{e}_\theta - \frac{\partial Y_{lm}^*(\theta, \varphi)}{\partial \theta} \mathbf{e}_\varphi\right], \tag{A1}$$

$$\begin{aligned}\{\nabla \times [j_l(pk_{01} r')\mathbf{X}_{lm}(\theta', \varphi')]\}^* &= -i[l(l+1)]^{1/2} \times \frac{j_l(pk_{01}r')}{r'} Y_{lm}^*(\theta', \varphi')\mathbf{e}_r + \frac{1}{r'} \frac{d[r' j_l(pk_{01}r')]}{dr'} \mathbf{e}_r \times \mathbf{X}_{lm}^*(\theta', \varphi') \\ &= -i[l(l+1)]^{1/2} \frac{j_l(pk_{01}r')}{r'} Y_{lm}^*(\theta', \varphi')\mathbf{e}_r + \frac{1}{r'} \frac{d[r' j_l(pk_{01}r')]}{dr'} \frac{-i}{[l(l+1)]^{1/2}} \times \frac{1}{\sin\theta'} \frac{\partial Y_{lm}^*(\theta', \varphi')}{\partial \varphi'} \mathbf{e}_\varphi + \frac{1}{r'} \frac{d[r' j_l(pk_{01}r')]}{dr'} \times \frac{-i}{[l(l+1)]^{1/2}} \frac{\partial Y_{lm}^*(\theta', \varphi')}{\partial \theta'} \mathbf{e}_\theta.\end{aligned} \tag{A2}$$

With these expressions the field components become

$$E_{2\theta}{}^{1} = 4\pi k^3 M n_2 \mu_1 \alpha_1 \frac{\exp(ik_2 r)}{k_2 r} \sum_{l,m} \frac{(-i)^{l+1} i}{[l(l+1)]^{1/2}} \times \Bigg\{ \frac{d_M(l)}{[l(l+1)]^{1/2}} \frac{1}{\sin\theta \sin\theta'} \times j_l(pk_{01}r') \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\varphi'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi} \times \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta - \frac{d_M(l)}{[l(l+1)]^{1/2}} \frac{1}{\sin\theta} j_l(pk_{01}r') \frac{\times\, \partial Y_{lm}{}^*(\theta', \varphi')}{\partial\theta'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi + \frac{id_E(l)}{kn_2} [l(l+1)]^{1/2} \frac{j_l(pk_{01}r')}{r'} Y_{lm}{}^*(\theta', \varphi') \times \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_r + \frac{id_E(l)}{kn_2[l(l+1)]^{1/2}} \times \frac{1}{\sin\theta'} \frac{1}{r'} \frac{\mathrm{d}[r_0 j_l(pk_{01}r')]}{\mathrm{d}r'} \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\varphi'} \times \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi + \frac{id_E(l)}{kn_2[l(l+1)]^{1/2}} \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \times \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\theta'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta \Bigg\}, \tag{A3}$$

$$E_{2\varphi}{}^{1} = -4\pi k^3 M n_2 \mu_1 \alpha_1 \frac{\exp(ik_2 r)}{k_2 r} \times \sum_{l,m} (-i)^{l+1} \frac{i}{[l(l+1)]^{1/2}} \Bigg\{ \frac{d_M(l)}{[l(l+1)]^{1/2}} \frac{1}{\sin\theta'} \times j_l(pk_{01}r') \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\varphi'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta} \times \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta - \frac{d_M(l)}{[l(l+1)]^{1/2}} j_l(pk_{01}r') \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\theta'} \times \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi - \frac{id_E(l)}{kn_2} [l(l+1)]^{1/2} \frac{1}{\sin\theta} \frac{j_l(pk_{01}r')}{r'} \times Y_{lm}{}^*(\theta', \varphi') \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi} \times \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_r - \frac{id_E(l)}{kn_2[l(l+1)]^{1/2}} \frac{1}{\sin\theta \sin\theta'} \frac{1}{r'} \times \frac{\mathrm{d}[r' j_l(pk_{01}r')] \partial Y_{lm}{}^*(\theta', \varphi')}{\mathrm{d}r' \quad \partial\varphi'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi} \times \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi - \frac{id_E(l)}{kn_2[l(l+1)]^{1/2}} \frac{1}{\sin\theta} \frac{1}{r'} \times \frac{\mathrm{d}[r' j_l(pk_{01}r')] \partial Y_{lm}{}^*(\theta', \varphi')}{\mathrm{d}r' \quad \partial\theta'} \times \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta \Bigg\}. \tag{A4}$$

Further, we use the addition theorem[12]

$$P_l(\cos\gamma) = \frac{4\pi}{2l+1} \sum_{m=-l}^{l} Y_{lm}{}^*(\theta', \varphi') Y_{lm}(\theta, \varphi) = \frac{4\pi}{2l+1} \sum_{m=-l}^{l} Y_{lm}{}^*(\theta, \varphi) Y_{lm}(\theta', \varphi'),$$

where $P_l(\cos\gamma)$ is the Legendre polynomial and $\cos\gamma = \cos\theta' \cos\theta + \sin\theta' \sin\theta \cos(\varphi' - \varphi)$. By partial differentiation of both parts of the formula we form the following relations:

$$S_{\varphi\varphi} = \frac{1}{\sin\theta' \sin\theta} \frac{\partial^2 P_l(\cos\gamma)}{\partial\varphi' \partial\varphi} = \frac{4\pi}{2l+1} \frac{1}{\sin\theta' \sin\theta} \times \sum_{m=-l}^{l} \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\varphi'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi},$$

$$S_{\theta\varphi} = \frac{1}{\sin\theta} \frac{\partial^2 P_l(\cos\gamma)}{\partial\theta' \partial\varphi} = \frac{4\pi}{2l+1} \frac{1}{\sin\theta} \sum_{m=-l}^{l} \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\varphi'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi},$$

$$S_\theta = \frac{\partial P_l(\cos\gamma)}{\partial\theta} = \frac{4\pi}{2l+1} \sum_{m=-l}^{l} Y_{lm}{}^*(\theta', \varphi') \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta},$$

$$S_{\varphi\theta} = \frac{1}{\sin\theta'} \frac{\partial^2 P_l(\cos\gamma)}{\partial\varphi' \partial\theta} = \frac{4\pi}{2l+1} \frac{1}{\sin\theta'} \sum_{m=-l}^{l} \frac{\partial Y_{lm}{}^*(\theta' \varphi')}{\partial\varphi'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta},$$

$$S_{\theta\theta} = \frac{\partial^2 P_l(\cos\gamma)}{\partial\theta' \partial\theta} = \frac{4\pi}{2l+1} \sum_{m=-l}^{l} \frac{\partial Y_{lm}{}^*(\theta', \varphi')}{\partial\theta'} \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\theta},$$

$$S_\varphi = \frac{1}{\sin\theta} \frac{\partial P_l(\cos\gamma)}{\partial\varphi} = \frac{4\pi}{2l+1} \frac{1}{\sin\theta} \sum_{m=-l}^{l} Y_{lm}{}^*(\theta', \varphi') \frac{\partial Y_{lm}(\theta, \varphi)}{\partial\varphi}.$$

Now the field components [Eqs. (B1) and (B2) below] can be rewritten without double sums:

$$E_{2\theta}{}^{1} = k^3 M n_2 \mu_1 \alpha_1 \frac{\exp(ik_2 r)}{k_2 r} \sum_{l=1}^{\infty} (-i)^{l+1} \frac{2l+1}{l(l+1)} \times \Big\{ i d_M(l) j_l(pK_1 r') S_{\varphi\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta - i d_M(l) j_l(pK_1 r') S_{\theta\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi - \frac{d_E(l)}{kn_2} l(l+1) \frac{j_l(pK_1 r')}{r'} S_\theta \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_r - \frac{d_E(l)}{kn_2} \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pK_1 r')]}{\mathrm{d}r'} S_{\varphi\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi - \frac{d_E(l)}{kn_2} \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pK_1 r')]}{\mathrm{d}r'} S_{\theta\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta \Big\},$$

$$E_{2\varphi}{}^{1} = -k^3 M n_2 \mu_1 \alpha_1 \frac{\exp(ik_2 r)}{k_2 r} \sum_{l=1}^{\infty} (-i)^{l+1} \frac{2l+1}{l(l+1)} \times \Big\{ i d_M(l) j_l(pK_1 r') S_{\varphi\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta - i d_M(l) j_l(pK_1 r') S_{\theta\theta} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi + \frac{d_E(l)}{kn_2} l(l+1) \frac{j_l(pK_1 r')}{r'} S_\varphi \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_r + \frac{d_E(l)}{kn_2} \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pK_1 r')]}{\mathrm{d}r'} S_{\varphi\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\varphi + \frac{d_E(l)}{kn_2} \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pK_1 r')]}{\mathrm{d}r'} S_{\theta\varphi} \mathbf{E}_1(\mathbf{r}') \cdot \mathbf{e}_\theta \Big\}.$$

Taking further programming into consideration, we can express the S functions as

$$S_{\varphi\varphi} = -\frac{\mathrm{d}^2 P_l}{\mathrm{d}x^2} \sin\theta' \sin\theta \sin^2\varphi' + \frac{\mathrm{d}P_l}{\mathrm{d}x} \cos\varphi_0,$$

$$S_{\theta\varphi} = \frac{\mathrm{d}^2 P_l}{\mathrm{d}x^2} \sin\theta' \sin\varphi' (-\sin\theta' \cos\theta + \cos\theta' \sin\theta \cos\varphi') + \frac{\mathrm{d}P_l}{\mathrm{d}x} \cos\theta' \sin\varphi',$$

$$S_{\varphi\theta} = \frac{\mathrm{d}^2 P_l}{\mathrm{d}x^2} \sin\theta \sin\varphi' (\cos\theta' \sin\theta - \sin\theta' \cos\theta \cos\varphi') - \frac{\mathrm{d}P_l}{\mathrm{d}x} \cos\theta \sin\varphi',$$

$$S_{\theta\theta} = \frac{\mathrm{d}^2 P_l}{\partial x^2} (\sin\theta' \cos\theta - \cos\theta' \sin\theta \cos\varphi') \times (\cos\theta' \sin\theta - \sin\theta' \cos\theta \cos\varphi') + \frac{\mathrm{d}P_l}{\partial x} (\sin\theta' \sin\theta + \cos\theta' \cos\theta \cos\varphi'),$$

$$S_\theta = \frac{\mathrm{d}P_l}{\mathrm{d}x} (-\cos\theta' \sin\theta + \sin\theta' \cos\theta \cos\varphi'),$$

$$S_\varphi = \frac{\mathrm{d}P_l}{\mathrm{d}x} \sin\theta' \sin\varphi',$$

where $x = \cos\gamma$ and the scattering plane is chosen at an angle $\varphi = 0$. From the recurrence relation for Legendre polynomials[15]

$$(2l+1) x P_l - (l+1) P_{l+1} - l P_{l-1} = 0,$$

we can find the derivatives

$$\frac{\mathrm{d}P_l}{\mathrm{d}x} = 1 \left[(2l-1) P_{l-1} + (2l-1) x \frac{\mathrm{d}P_{l-1}}{\mathrm{d}x} - (l-1) \frac{\mathrm{d}P_{l-2}}{\mathrm{d}x} \right] / l,$$

$$\frac{\mathrm{d}^2 P_l}{\mathrm{d}x^2} = l \left[2(2l-1) \frac{\mathrm{d}P_{l-1}}{\mathrm{d}x} + (2l-1) x \frac{\mathrm{d}^2 P_{l-1}}{\mathrm{d}x^2} - (l-1) \frac{\mathrm{d}^2 P_{l-2}}{\mathrm{d}x^2} \right] / l.$$

Taking into account the initial values $P_0 = 1$, $P_1 = x$, $\mathrm{d}P_0/\mathrm{d}x = 0$, $\mathrm{d}P_1/\mathrm{d}x = 1$, $\mathrm{d}^2P_0/\mathrm{d}x^2 = 0$, and $\mathrm{d}^2P_1/\mathrm{d}x^2 = 0$, we can calculate all the S functions sequentially from $l = 1$ to $l = l_{\max}$.

Appendix B: Evaluation of Field Components for Backscattering Geometry

After substitution of expressions (26)–(28) into components of scattered field $\mathbf{E}_2{}^1(\mathbf{r}')$ we have

$$E_{2\theta} = \frac{\exp(ik_2 r)}{k_2 r} \sum_{l=1}^{\infty} (-i)^{l+1} (-1)^l \frac{i(2l+1)^{1/2}}{2\sqrt{4\pi}} \times \left\{ i[c_M(l,1) + c_M(l,-1)] + \frac{c_E(l,1) - c_E(l,-1)}{n_2} \right\},$$

$$E_{2\varphi} = \frac{\exp(ik_2 r)}{k_2 r} \sum_{l=1}^{\infty} (-i)^{l+1} (-1)^l \frac{i(2l+1)^{1/2}}{2\sqrt{4\pi}} \times \left[c_M(l,1) - c_M(l,-1) - i \frac{c_E(l,1) + c_E(l,-1)}{n_2} \right]. \quad \text{(B1)}$$

Further simplification can be made if we combine complex conjugates of vector spherical harmonics that carry equal subscripts l and express these pairs in spherical coordinates:

$$\mathbf{X}_{l,1}{}^* + \mathbf{X}_{l,-1}{}^* = \frac{2(2l+1)^{1/2}}{l(l+1)\sqrt{4\pi}} \left[\mathbf{e}_\varphi \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} \sin\varphi' - \mathbf{e}_\theta \frac{P_l{}^1(\theta')}{\sin\theta'} \cos\varphi' \right],$$

$$\mathbf{X}_{l,1}{}^* - \mathbf{X}_{l,-1}{}^* = \frac{2i(2l+1)^{1/2}}{l(l+1)\sqrt{4\pi}} \left[\mathbf{e}_\varphi \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} \cos\varphi' + \mathbf{e}_\theta \frac{P_l{}^1(\theta')}{\sin\theta'} \sin\varphi' \right],$$

$$\nabla \times [j_l(pk_{01}r')(\mathbf{X}_{l,1}{}^* + \mathbf{X}_{l,-1}{}^*)] = -\frac{2(2l+1)^{1/2}}{l(l+1)\sqrt{4\pi}} \times \left\{ \mathbf{e}_r l(l+1) \frac{j_l(pk_{01}r')}{r'} P_l{}^1(\theta')\sin\varphi' + \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \left[\mathbf{e}_\theta \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} \sin\varphi' + \mathbf{e}_\varphi \frac{P_l{}^1(\theta')}{\sin\theta'} \cos\varphi' \right] \right\},$$

$$\nabla \times [j_l(pk_{01}r')(\mathbf{X}_{l,1}{}^* - \mathbf{X}_{l,-1}{}^*)] = -\frac{2i(2l+1)^{1/2}}{l(l+1)\sqrt{4\pi}} \times \left\{ \mathbf{e}_r l(l+1) \frac{j_l(pk_{01}r')}{r'} P_l{}^1(\theta')\cos\varphi' + \frac{1}{r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \left[\mathbf{e}_\theta \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} \cos\varphi' - \mathbf{e}_\varphi \frac{P_l{}^1(\theta')}{\sin\theta'} \sin\varphi' \right] \right\}.$$

In both cases of incident wave orientation the components of scattered field will then look as follows:

$$E_{2\theta}{}^{1OX} = -E_{2\varphi}{}^{1OY} = E_0 \frac{\exp(ik_2 r)}{r} k^2 M \mu_1 \alpha_1 \times \left\{ \cos^2\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} \times d_M(l) A_\theta j_l(pk_{01}r') \frac{P_l{}^1(\theta')}{\sin\theta'} - \sin^2\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} d_M(l) A_\varphi j_l(pk_{01}r') \times \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} - \cos^2\varphi' \sum_{l=1}^{\infty} i^l (2l+1) i d_E(l) A_r \frac{j_l(pk_{01}r')}{k_2 r'} P_l{}^1(\theta') - \cos^2\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)i}{l(l+1)} d_E(l) A_\theta \times \frac{1}{k_2 r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} + \sin^2\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)i}{l(l+1)} d_E(l) A_\varphi \times \frac{1}{k_2 r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \frac{P_l{}^1(\theta')}{\sin\theta'} \right\}, \tag{B2}$$

$$E_{2\varphi}{}^{1OX} = -E_{2\theta}{}^{1OY} = E_0 \frac{\exp(ik_2 r)}{r} k^2 M \mu_1 \alpha_1 \left\{ -\sin\varphi' \cos\varphi' \times \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} k d_M(l) A_\theta j_l(pk_{01}r') \times \frac{P_l{}^1(\theta')}{\sin\theta'} - \sin\varphi' \cos\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)}{l(l+1)} \times k d_M(l) A_\varphi j_l(pk_{01}r') \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} + \sin\varphi' \cos\varphi' \sum_{l=1}^{\infty} i^l (2l+1) i d_E(l) A_r \frac{j_l(pk_{01}r')}{k_2 r'} P_l{}^1(\theta') + \sin\varphi' \cos\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)i}{l(l+1)} \times d_E(l) A_\theta \frac{1}{k_2 r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \frac{\mathrm{d}P_l{}^1(\theta')}{\mathrm{d}\theta'} + \sin\varphi' \cos\varphi' \sum_{l=1}^{\infty} i^l \frac{(2l+1)i}{l(l+1)} \times d_E(l) A_\varphi \frac{1}{k_2 r'} \frac{\mathrm{d}[r' j_l(pk_{01}r')]}{\mathrm{d}r'} \frac{P_l{}^1(\theta')}{\sin\theta'} \right\}. \tag{B3}$$

Substituting Riccati–Bessel functions for the spherical Bessel functions and using expressions (31), we can write the field components as

$$E_{2\theta}{}^{1OX} = -E_{2\varphi}{}^{1OY} = E_0 \frac{\exp(ik_2 r)}{r} k^2 M \mu_1 \alpha_1 [B_1(r', \theta')\cos^2\varphi' - B_2(r', \theta')\sin^2\varphi' - iB_3(r', \theta')\cos^2\varphi' - iB_4(r', \theta')\cos^2\varphi' + iB_5(r', \theta')\sin^2\varphi'],$$

$$E_{2\varphi}{}^{1OX} = -E_{2\theta}{}^{1OY} = E_0 \frac{\exp(ik_2 r)}{r} k^2 M \mu_1 \alpha_1 [-B_1(r', \theta') - B_2(r', \theta') + iB_3(r', \theta') + iB_4(r', \theta') + iB_5(r', \theta')]\sin\varphi' \cos\varphi'.$$

After integration of these expressions over angle φ', we obtain the final expressions, Eqs. (29) and (30).

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