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Modified P_N for Correlation Transfer in One-Dimensional Scattering and Absorbing Media

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Previously, correlation transfer (CT) theory was developed with the ability to handle dynamic light scattering measurements from single scattering to highly multiple scattering. Because of the lengthy computational time required to obtain exact results for the CT equation, the modified P_N approximation has been applied to handle any order of Legendre approximation. Herein, numerical solutions of the modified P_N approximation are given for backscattering and transmission at optical thicknesses of 1, 5, 10, and 25. Fresnel reflective boundaries with a collimated incident laser beam have been used to provide results to compare with exact solutions. The significantly reduced computational time and reasonable agreement with the exact solution make this approximation an effective technique for quickly obtaining CT equation solutions.

Nomenclature

A_n = location dependent coefficient of Legendre expansion for intensity
c = multiple scattering intensity correlation function, $W^2/m^4 \cdot rad^2$
c_0 = speed of light, m/s
D_0 = diffusion constant of particles in the medium, m^2/s
d = diameter of particle, m
$d\Omega$ = differential solid angle around the direction, Ω
E = magnitude of electric field, N/C
f_m = phase function Legendre expansion coefficient
G = electric field correlation function
G^m = multiple scattering field correlation function
G_i^m = incident multiple scattering field correlation
g^1 = single scattering field correlation function
I = intensity of radiation, $W/m^2 \cdot sr$
k = magnitude of the free medium's wave vector, m^{-1}
k_B = Boltzmann constant, J/K
k_0 = magnitude of the incident wave vector, m^{-1}
L = optical coordinate
L_0 = optical thickness
N = highest order of Legendre expansion
n = refractive index
P_i = Legendre polynomial of order i
r = spatial position vector, m
S = source function, W/m^2
s = parameter used in computing Fresnel's reflection
T = absolute temperature, K
T_e = total experimental time for one run, s
t = time, s
x_i = ith coefficient of Legendre polynomial expansion
γ = experimental proportionality constant
η = solvent viscosity, $N \cdot s/m^2$
Θ = scattering angle
κ = absorption coefficient, m^{-1}
λ = wavelength, m
μ = cosine of the polar angle, $\cos\theta$
ρ = reflectivity coefficient
σ = scattering coefficient, m^{-1}
τ = correlation delay time, s
τ_0 = characteristic delay time, $1/D_0k_0^2$, s
Φ = scattering phase function
$\hat{\mathbf{\Omega}}'$ = unit vector in scattered direction
$\hat{\mathbf{\Omega}}$ = unit vector in incident direction
ω = single scattering albedo
ω_e = effective albedo

Introduction

THE classical P_N (spherical harmonics) method is one of the preferred approximations used in radiative transfer. Viskanta and Menguc[1] discussed the applications of the classical P_N method in combustion. They indicate that the P_1 approximation is accurate for flux in media having an optical thickness greater than 2.0. Although P_1 is not accurate for small optical thickness, this defect can be improved by using higher-order moments of intensity such as the P_3 approximation. Howell[2] wrote a general review of approximate methods that are commonly used in radiative transfer. According to that review, the classical P_N approximation is usually inaccurate for the prediction of intensity. To achieve a good approximation for the intensity, a very high-order classical P_N approximation must be used.

To obtain better results from the P_N approximation, improvement can be achieved by substituting the intensity calculated from the classical P_N method into the source function equation for radiative transfer, and then recalculating the intensity and flux. In this regard, Modest[3,4] and Modest and Stevens[5] applied this improvement to the P_1 approximation for two-dimensional nonscattering media in the radiative equation. Also, Modest[6,7] extended this improvement to linearly anisotropic scattering in three-dimensional media. However, all of these improvements were done for the P_1 approximation and for flux calculations only. No attention was focused on higher-order approximations or on intensity calculations.

Recently, a series of investigations were performed at Los Alamas National Laboratory on the P_1 solution for flux. Olson et al.[8] compared the P_1 approximation to other diffusion approximation techniques. They also investigated the application of the P_1 solution in optically thin media by modifying the velocity of propagation. Morel[9] developed an asymptotic solution of the transport equation

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in the diffusion limit as a reference and compared the Olson et al.[8] P_1 velocity modification (termed $P_{1/3}$) and two other diffusion approximations to it. Morel[9] studied flux in the equilibrium diffusion limit with no scattering. Simmons and Mihalas[10] investigated $P_{1/3}$ for flux with a correction for dilute samples, but they did not present any results.

To date, most of the research on the P_N method has been concerned with its application in the prediction of the radiative flux, and reports using the P_N approximation to obtain intensity are limited. Benassi et al.[11] developed the solution for flux and intensity in an emitting medium with both specular and diffuse reflection at the boundaries using the classical P_N method. They used a special form for the intensity, as had been done by Garcia and Siewert.[12] Benassi et al.[11] compared their heat flux results with the exact solution provided by Siewert et al.,[13] who were using the F_N method. They did not present results for intensity. In another paper, Benassi et al.[14] presented very accurate intensity and heat flux results by using high-order ($N > 40$) classical P_N approximations. In their study, one-dimensional plane parallel radiative transfer with a collimated boundary condition was considered. In another study, Siewert and Thomas[15] determined the particular solution for the form of intensity expansion from Garcia and Siewert[12] using general reflective boundary conditions. Later, Siewert[16,17] developed the same spherical harmonic approximation for an inverse source problem and nongray radiative transport. However, the solution for intensity based on the Garcia and Siewert[12] expansion form requires a series of mathematical computations, which causes the spherical harmonics method to become a very complicated solution procedure. McCormick[18] discussed the use of specialized polynomials in spherical harmonics, and Barichello et al.[19] developed the P_N approximation with reflecting boundaries and an internal source and discussed the singularity of the intensity due to the incident delta function model. They used Fourier decomposition to obtain better results around singular points. Muldashev et al.[20] applied the P_N approximation in an atmosphere-to-Earth's-surface system. They used a correction function for smoothing intensity oscillations in lower-order spherical harmonics. Their P_{33} solution compared well to the standard P_{499} and discrete ordinate results for intensity of radiance. Recently, Lyapustin and Muldashev[21] applied the previous smoothing correction function to their atmospheric optical transfer function with diffuse reflectance and albedo variation. They presented a one-dimensional solution with a diffuse boundary condition. They did not compare their numerical results to other solutions.

Wilson and Sen[22] modified the classical spherical harmonics solution using a different expansion of the intensity in Legendre polynomials. They solved for the first-order approximation using the Rayleigh phase function and showed improvement of the intensity calculation over the third approximation of Chandrasekhar's[23] Gausian quadrature method. Later, Wan et al.[24,25] improved this modification. They presented solutions for the first-order approximation with Rayleigh and linearly anisotropic phase functions for the first and second moment of the intensity. Karanjai and Biswas[26] expanded the same solution for the second-order approximation, but they did not present any results.

In this paper, we have developed an improvement for the P_N approximation for the intensity in the same general manner as was done by Modest[6,7] for the flux using the P_1 approximation. In addition, we have included the index of refraction at the boundaries and considered absorbing and scattering media. We have applied this technique to the correlation transfer (CT) equation. The CT equation, which was proposed by Ackerson et al.,[27] is based on the fluctuation of the radiative intensity with time (dynamic light scattering), and it is similar to the radiative transport (RT) equation. In this dynamic light scattering technique, the intensity fluctuation in fluid/particle suspensions, due to Brownian motion of the particles, is being studied. The fluctuation is compared (correlated) with itself, and shifted by user-selected delay times. These time displaced intensity functions are multiplied together and compared over large amounts of sample times to form an intensity correlation function. The comparison of these correlation functions with theory can be used to determine the physical properties, for example, particle sizes and fluid viscosity, of the fluid/particle suspensions being probed.

The CT equation was proposed to study the correlation function from a multiple scattering medium. Because the mathematical form of the CT equation is very similar to that of the RT equation, the theoretical solution techniques applied in radiative transfer can be applied directly to the CT equation. Reguigui et al.[28] have outlined the step-by-step development of the CT equation and have shown good comparison of the CT equation's exact solution with experimental data for optical thickness as low as five in both transmission and backscattering geometries. They also improved the numerical results over those of Ackerson et al.[27] by approximating the single scattering correlation function g^1, which is part of the effective phase function in the CT equation, as a Legendre series. Ackerson et al.[27] used a preaveraging method for g^1 to obtain their numerical results. The improved results of Reguigui et al.[28] compared well with experimental data for semi-infinite media over a longer nondimensional delay time (τ/τ_0) period than that presented by Ackerson et al.[27]

Dorri-Nowkoorani et al.[29] presented a variety of experimental correlation functions for scattering media and compared them to the exact solution of the CT equation. They also proposed a methodology for characterizing the particle size in the multiple scattering regimes. Because the numerical calculations to obtain the exact solution results have very long execution times, especially for high optical thickness or when index of refraction effects are included, exact procedures are not very convenient for real-time solution of the CT equation, especially for particle size characterization. Here, we will compare the numerical results (and calculation times) from the improved P_N approximation to the exact solution of the three-term Legendre expansion of the single scattering correlation function g^1.

Theoretical Background

The description of the time dependency of the dynamic light scattering technique can be found in detail in Ref. 30. Here, we give a very brief review of a few basic concepts. In a fluid/particle suspension, the particles experience Brownian motion, and their positions are continually changing. The scattered electric field, which is a function of particle position, is also continuously changing. Because the intensity is proportional to the square of the electric field,[31] intensity is also fluctuating in time. The dynamic information of interest is contained in the fluctuations, and the most efficient way to analyze the intensity fluctuations is to average the product of the signal from a detector and a (time) delayed version of the signal as a function of that delay time τ. This is known as autocorrelation, and the intensity autocorrelation function is defined as follows[30]:

$$c(\tau) = \langle I(t)I(t+\tau)\rangle = \lim_{T_e\to\infty}\frac{1}{T_e}\int_0^{Te} I(t)I(t+\tau)\,dt \propto \{1+\gamma[G(\tau)]^2 \qquad (1)$$

where the intensity I has, in general, different values at time t and $t+\tau$. The angle brackets indicate an ensemble average over the particles present, approximated by an integral of intensities. T_e represents the total experimental duration time, over which the product of the intensity with delayed versions of itself is averaged. $G(\tau)$ is known as the electric field correlation function:

$$G(\tau) = \langle E(t)E^*(t+\tau)\rangle \qquad (2)$$

where E^* is the complex conjugate of the electric field. The intensity correlation function of Eq. (1) is measured experimentally by commercially available correlator hardware and/or software that multiplies the shifted intensity signals together and integrates the results.[31] For very large delay time, the autocorrelation function decays and is equal to the square of the average intensity, that is, $\langle I(t)\rangle^2$.

Single Scattering

For very dilute and monodisperse samples, the single scattering correlation function is[30]

$$g^1(k,\tau) = \exp\left(-D_0 \boldsymbol{k}^2 \tau\right) = \exp\left[-2D_0 k_0^2 \tau(1-\cos\Theta)\right] \quad (3)$$

and it depends on the scattering wave vector $\boldsymbol{k}$ with magnitude $k = 2\pi n/\lambda$ and the diffusion constant D_0. The relation $\boldsymbol{k}^2 = 2k_0^2(1-\cos\Theta)$ has been used in Eq. (3), where Θ is the scattering angle measured from the forward direction. The diffusion constant D_0 is given by the Stokes–Einstein relation for spherical particles (see Ref. 32):

$$D_0 = k_B T/3\pi\eta d \quad (4)$$

where d is the diameter of a particle suspended in the sample. Another useful parameter, which is used to nondimensionalize delay time, is the correlation delay time constant τ_0:

$$\tau_0 = 1/D_0 k_0^2 \quad (5)$$

Multiple Scattering

Equation (3) is only valid for dilute samples. In concentrated samples, the correlation function looks different from Eq. (3). In that case, the integro–differential equation[28]

$$\left(\frac{1}{c_0}\right)\frac{\partial G^m(\boldsymbol{r},\boldsymbol{\Omega},t,\tau)}{\partial t} + \Omega\cdot\nabla G^m(\boldsymbol{r},\boldsymbol{\Omega},t,\tau) + \kappa G^m(\boldsymbol{r},\boldsymbol{\Omega},t,\tau) + \sigma G^m(\boldsymbol{r},\boldsymbol{\Omega},t,\tau) = \frac{\sigma}{4\pi}\int_{4\pi} G^m(\boldsymbol{r},\boldsymbol{\Omega}',t,\tau) g^1(k',\tau)\Phi(\boldsymbol{\Omega},\boldsymbol{\Omega}')\,\mathrm{d}\Omega' \quad (6)$$

has to be solved for G^m, the multiple scattering correlation function subject to the appropriate boundary conditions of the problem. The integration over all directions $\boldsymbol{\Omega}'$ takes into account the correlation scattered from other directions into the direction of interest. The kernel is made up of two functions: Φ is the form factor (phase function), and g^1 is the single scattering function that governs the transport of correlation. The first two terms on the left side denote the temporal and spatial propagation, respectively. The next two terms are the absorption and scattering of correlation. This CT equation is mathematically the same as the RT equation of Siegel and Howell[32]:

$$\left(\frac{1}{c_0}\right)\frac{\partial I(\boldsymbol{r},\boldsymbol{\Omega},t)}{\partial t} + \Omega\cdot\nabla I(\boldsymbol{r},\boldsymbol{\Omega},t) + \kappa I(\boldsymbol{r},\boldsymbol{\Omega},t) + \sigma I(\boldsymbol{r},\boldsymbol{\Omega},t) = \frac{\sigma}{4\pi}\int_{4\pi} I(\boldsymbol{r},\boldsymbol{\Omega}',t)\Phi(\boldsymbol{\Omega},\boldsymbol{\Omega}')\,\mathrm{d}\Omega' \quad (7)$$

except for $g^1(k',\tau)$ under the integral.

The one-dimensional form of Eq. (6) for a scattering and absorbing medium and assuming azimuthal symmetry is

$$\mu\frac{\mathrm{d}G^m(L,\mu,\tau)}{\mathrm{d}L} + G^m(L,\mu,\tau) = \frac{\omega}{2}\int_{-1}^{1} G^m(L,\mu',\tau) g^1(k',\tau)\Phi(\mu,\mu')\,\mathrm{d}\mu' \quad (8)$$

where L is the optical coordinate that is equal to

$$\int_0^z (\sigma+\kappa)\,\mathrm{d}z'$$

where ω is the single scattering albedo defined as $\sigma/(\sigma+\kappa)$. The solution of Eq. (8) can be found as[30]

$$G^m(L,\mu,\tau) = G_i^m(L,\mu,\tau)e^{-L/\mu} + \left(\frac{\omega}{2\mu}\right)\int_0^L\int_{-1}^{1} G^m(L',\mu',\tau) g^1(k',\tau)\Phi(\mu,\mu') \times \exp\left(\frac{|L-L'|}{\mu}\right)\mathrm{d}\mu'\,\mathrm{d}L' \quad (9)$$

where G_i^m is the incident multiple scattering field correlation. Equation (9) can be solved by the same methods as used for a standard RT equation with an extra term, the singly scattered correlation function g^1 under the integral. In the case of using the isotropic scattering assumption [$\Phi(\mu,\mu') = 1.0$] when applying RT solution techniques in finding G^m, one can expand g^1 in a series of Legendre polynomials[28]:

$$g^1(k,\tau) = \omega_e \sum_{i=0}^{M} x_i P_i(\cos\Theta) \quad (10)$$

In this case, x_0 is 1.0 and

$$\omega_e = (2\tau/\tau_0)^{-1}\exp(-2\tau/\tau_0)\sinh(2\tau/\tau_0) \quad (11)$$

where ω_e is the effective albedo and, in the case of a purely scattering medium ($\omega = 1.0$), ω is replaced by ω_e. The x_i are the Legendre expansion coefficients and are given by Reguigui et al.[28] for a second-order expansion.

More detailed information about the CT equation can be found in the work of Ackerson et al.[27] and Reguigui et al.[28]

Classical P_N Approximation

This approximation has been known to apply for the RT equation [Eq. (7)]. Because the RT equation is the same as the CT equation [Eq. (6)] except for $g^1(k',\tau)$ under the integral, we provide a brief summary of the development for intensity. We will use this development to provide a comparison for the modified P_N and then apply the results of that development to correlation.

The general transport equation for a one-dimensional absorbing and scattering medium is written as[33]

$$\mu\frac{\partial I(L,\mu)}{\partial L} + I(L,\mu) = S(L,\mu) = \frac{\omega}{2}\int_{-1}^{1} I(L,\mu')\Phi(\mu,\mu')\,\mathrm{d}\mu' \quad (12)$$

The scattering phase function $\Phi(\mu,\mu')$ can be represented as a series of Legendre polynomials,[33]

$$\Phi(\mu,\mu') = \sum_{m=0}^{N}(2m+1) f_m P_m(\mu) P_m(\mu') \quad (13)$$

where the coefficients f_m are specified depending on the anisotropic scattering in a given medium.

For the P_N approximation, the intensity $I(L,\mu)$ is also expanded in a series of Legendre polynomials,[33]

$$I(L,\mu) = \frac{1}{4\pi}\sum_{m=0}^{N}(2m+1) P_m(\mu) A_m(L) \quad (14)$$

When Eqs. (13) and (14) are put into Eq. (12), and integral and recurrence relations for Legendre polynomials[34] are used, the following relationship among the $A_m(L)$ can be found:

$$(m+1)\frac{\mathrm{d}A_{m+1}(L)}{\mathrm{d}L} + m\frac{\mathrm{d}A_{m-1}(L)}{\mathrm{d}L} + (2m+1)(1-\omega f_m)A_m(L) = 0 \quad (15)$$

This differential equation can be solved for the $A(L)$ by using numerical techniques for two-point boundary value problems.[35] Applying this solution technique coupled with the finite difference method produces a matrix with elements that consist of the $A_m(L)$ and boundary values. Here, Marshak's boundary conditions (see Ref. 33) are used, and they are

$$\int_0^1 I^+(0,\mu)\mu^{2i-1}\,\mathrm{d}\mu = \int_0^1 I_1^+(\mu)\mu^{2i-1}\,\mathrm{d}\mu = I_1 \quad (16a)$$

$$\int_0^1 I^-(L_0,\mu)\mu^{2i-1}\,\mathrm{d}\mu = \int_0^1 I_2^-(\mu)\mu^{2i-1}\,\mathrm{d}\mu = I_2 \quad (16b)$$

where $i = 1, 2, 3, \ldots, (N+1)/2$ and $I_1^+(\mu)$ and $I_2^-(\mu)$ are the boundary conditions at the $L = 0$ and L_0 locations for the medium, respectively.

The intensities $I^+(0, \mu)$ and $I^-(L_0, \mu)$ can be written using Eq. (14), but to reduce the size of the matrix created when solving Eq. (15), the following development will be applied to Eq. (15).

The odd and even numbers of m can be separated in Eq. (14) such that the intensity $I(L, \mu)$ is

$$I(L, \mu) = \frac{1}{4\pi} \sum_{\substack{m=0 \\ m=\text{even}}}^{N} (2m+1) P_m(\mu) A_m(L) + \frac{1}{4\pi} \sum_{\substack{m=1 \\ m=\text{odd}}}^{N} (2m+1) P_m(\mu) A_m(L) \tag{17}$$

By the substitution of $2k$ for m in the first summation and $2k+1$ for m in the second summation on the right-hand side of Eq. (17), and by the consideration only odd orders in solving the P_N formulation, the two summations in Eq. (17) can be combined and reduced to

$$I(L, \mu) = \frac{1}{4\pi} \sum_{k=0}^{(N-1)/2} [(4k+1) P_{2k}(\mu) A_{2k}(L) + (4k+3) P_{2k+1}(\mu) A_{2k+1}(L)] \tag{18}$$

$A_m(L)$ can be found as a function of $A'_{m+1}(L)$ and $A'_{m-1}(L)$ (prime indicates the derivative) from Eq. (15) and the derivatives can be substituted for even values of $A_m(L)$ in Eq. (18). Then the intensity equation becomes

$$I(L, \mu) = \frac{1}{4\pi} \sum_{k=0}^{(N-1)/2} \left[-\frac{(2k+1) A'_{2k+1}(L) + 2k A'_{2k-1}(L)}{(1 - \omega f_{2k})} P_{2k}(\mu) + (4k+3) P_{2k+1}(\mu) A_{2k+1}(L) \right] \tag{19}$$

When the closed-form representation of Legendre polynomials[34] is applied, Eq. (19) becomes

$$I(L, \mu) = \frac{1}{4\pi} \sum_{k=0}^{(N-1)/2} \left[-\frac{(2k+1) A'_{2k+1}(L) + 2k A'_{2k-1}(L)}{2^{2k}(1 - \omega f_{2k})} \times \sum_{r=0}^{k} (-1)^r \frac{(4k-2r)!}{(2k-r)! r! (2k-2r)!} \mu^{2k-2r} + \frac{(4k+3)}{2^{2k+1}} A_{2k+1}(L) \times \sum_{r=0}^{k} (-1)^r \frac{(4k-2r+2)!}{(2k-r+1)! r! (2k-2r+1)!} \mu^{2k-2r+1} \right] \tag{20}$$

where r is equal to positive integers. The equations for the boundary conditions can be found by inserting the intensity equation of Eq. (20) in the boundary conditions of Eqs. (16a) and (16b) and integrating over μ.

When Eq. (15) is solved for $A_m(L)$, its derivative is taken with respect to L, and then it is substituted back in Eq. (15) for $A'_{m+1}(L)$ and $A'_{m-1}(L)$, the recurrence relation for the odd subscripted $A_m(L)$ can be found, that is,

$$\frac{(m+1)(m+2)(1 - \omega f_{m-1})}{2m+3} \frac{\mathrm{d}^2 A_{m+2}(L)}{\mathrm{d}L^2} + \left[\frac{(m+1)^2 (1 - \omega f_{m-1})}{2m+3} + \frac{m^2 (1 - \omega f_{m+1})}{2m-1} \right] \frac{\mathrm{d}^2 A_m(L)}{\mathrm{d}L^2} + \frac{m(m-1)(1 - \omega f_{m+1})}{2m-1} \frac{\mathrm{d}^2 A_{m-2}(L)}{\mathrm{d}L^2} - (2m+1)(1 - \omega f_{m-1})(1 - \omega f_m)(1 - \omega f_{m+1}) A_m(L) = 0 \tag{21}$$

where $m = 1, 3, 5, \ldots, N$. In this recurrence relation, when m is equal to N, the parameters with $(m+1)$ should be zero, and f_{m+1} should also be zero.

The finite difference technique can be applied to Eqs. (21) to produce a matrix for calculating $A_m(L)$, and intensity is directly related to the $A_m(L)$ by either Eq. (19) or (20). This is the derivation of the classical P_N approximation. Next, we will show how the approximation can be improved.

Modified P_N Approximation

For $0 \le \mu \le 1.0$, the analytical solution of Eq. (12) is[33]

$$I^+(L, \mu) = I_1^+(\mu) \exp\left(-\frac{L}{\mu}\right) + \int_0^L S(L', \mu) \exp\left[-\frac{(L - L')}{\mu}\right] \frac{\mathrm{d}L'}{\mu} \tag{22a}$$

$$I^-(L, \mu) = I_2^-(\mu) \exp\left[-\frac{(L_0 - L)}{\mu}\right] + \int_L^{L_0} S(L', \mu) \exp\left[-\frac{(L' - L)}{\mu}\right] \frac{\mathrm{d}L'}{\mu} \tag{22b}$$

For the P_N approximation, the source function $S(L, \mu)$, which is defined in Eq. (12), is found by substituting Eqs. (13) and (14) into the right-hand side of Eq. (12), yielding

$$S(L, \mu) = \frac{\omega}{4\pi} \sum_{m=0}^{N} (2m+1) A_m(L) f_m P_m(\mu) \tag{23}$$

Thus, the P_N approximation can be improved by substituting Eq. (23) into Eqs. (22) and solving for the backscattered or transmitted intensities. Because the $A_m(L)$ remain inside the integral, and a closed-form solution of $A_m(L)$ is not available for higher orders of the P_N approximation ($N > 1$), these integral equations for $A_m(L)$ have to be solved numerically using, in this case, Guass–Legendre quadrature for the integration.

Boundary Conditions

The boundary conditions $I_1^+(\mu)$ and $I_2^-(\mu)$ in Eq. (20) can be defined as collimated incident light with an index of refraction change at both top and bottom interfaces. In this case, they are formulated as[36]

$$I_1^+(\mu) = \mu_{1o} I_{1o} \frac{[1 - \rho(\mu_{1o}, 1/n_1)]}{\left[1 - \left(1 - \mu_{1o}^2\right)/n_1^2\right]^{\frac{1}{2}}} \delta \left\{ \mu - \left[1 - \frac{\left(1 - \mu_{1o}^2\right)}{n_1^2} \right]^{\frac{1}{2}} \right\} + I^-(0, \mu) \rho(\mu, n_1) \tag{24a}$$

$$I_2^+(\mu) = \mu_{2o} I_{2o} \frac{[1 - \rho(\mu_{2o}, 1/n_2)]}{\left[1 - \left(1 - \mu_{2o}^2\right)/n_2^2\right]^{\frac{1}{2}}} \delta \left\{ \mu - \left[1 - \frac{\left(1 - \mu_{2o}^2\right)}{n_2^2} \right]^{\frac{1}{2}} \right\} + I^+(L, \mu) \rho(\mu, n_2) \tag{24b}$$

where subscripts 1 and 2 refer to top and bottom boundaries, respectively, and subscript o denotes the outside of the medium. The reflectivity ρ is calculated from Fresnel's reflection equation as[33]

$$\rho(\mu, n) = \frac{1}{2} \left[\left(\frac{s - \mu}{s + \mu} \right)^2 + \left(\frac{s - \mu/n^2}{s + \mu/n^2} \right)^2 \right] \tag{25}$$

where $s = \sqrt{[1/n^2 - (1 - \mu^2)]}$. Substituting Eqs. (24) into (16) yields

$$I_1 = \mu_{1o} I_{1o}\left[1-\rho\left(\mu_{1o},\frac{1}{n_1}\right)\right]\left[1-\frac{\left(1-\mu_{1o}^2\right)}{n_1^2}\right]^{i-1} + \int_0^1 I^-(0,\mu)\rho(\mu,n_1)\mu^{2i-1}\,\mathrm{d}\mu \tag{26a}$$

$$I_2 = \mu_{2o} I_{2o}\left[1-\rho\left(\mu_{1o},\frac{1}{n_2}\right)\right]\left[1-\frac{\left(1-\mu_{2o}^2\right)}{n_2^2}\right]^{i-1} + \int_0^1 I^+(L,\mu)\rho(\mu,n_2)\mu^{2i-1}\,\mathrm{d}\mu \tag{26b}$$

where $i = 1, 2, 3, \ldots, (N+1)/2$.

The intensities, $I^-(0,\mu)$ and $I^+(L,\mu)$, are unknown. Thus, by the initial assumptions of some values and by the use of an iteration technique, the results will be obtained if refractive indexes other than 1.0 are selected for n_1 and n_2.

As mentioned before, the development of the modified P_N approximation for intensity can be applied to the correlation function due to the similarity of RT [Eq. (12)] and CT [Eq. (8)]. In this case, $I(L,\mu)$ and $\Phi(\mu,\mu')$ are replaced by $G^m(L,\mu,\tau)$ and $g^1(k,\tau)\Phi(\mu,\mu')$, respectively. Accordingly, f_m in Eq. (13) and $A_m(L)$ in Eq. (14) will change to $f_m(\tau)$ and $A_m(L,\tau)$.

Results and Discussion

For the plots discussed herein, the results from correlation transfer are plotted as the natural logarithm of the correlation function vs the square root of nondimensional delay time (τ/τ_0) because there is a near linear dependency between these two parameters for infinite optical thickness.[27] Examining these types of plots allows one to more easily analyze the data because the degree of fit/deviation from a straight line is clearly discernible.

Obtaining the exact results from the CT equation requires significant computational time, especially if index of refraction and a Legendre expansion for single scattering g^1 are included in the calculations. As the required optical thickness increases, the execution time for the program for the exact solution increases, and such lengthy solution times will not be suitable for particle sizing when a rapid turn around for the results is desired. Because the CT equation is similar to the RT equation, and expansion of the single scattering g^1 into additional terms in the Legendre series can improve the theoretical results, the modified P_N technique may be used to solve the CT equation, if the P_N solution can be shown to compare well with the exact solution. For simplicity, isotropic scattering $[\Phi(\mu,\mu')=1]$ correlation function results are presented here.

In the following results, the single scattering g^1 in Eq. (9) has been expanded to three terms in a Legendre series ($i = 0$, 1, and 2), whereas the correlation function G^m has not been approximated. This is called the exact three-term case. In the P_N approximation, both the correlation function G^m and single scattering g^1 were expanded to the same $N+1$ terms in a Legendre series. This means that P_1 has two terms, the zeroth and the first order, P_3 has four terms, the zeroth, first, second, and third order, and so on.

Figures 1–8 present a comparison of the exact unit refractive index solution[37] for the three-term g^1 expansion in a Legendre series to the numerical results using the modified P_1, P_3, P_5, and P_7 approximations for backscattering and transmission at optical

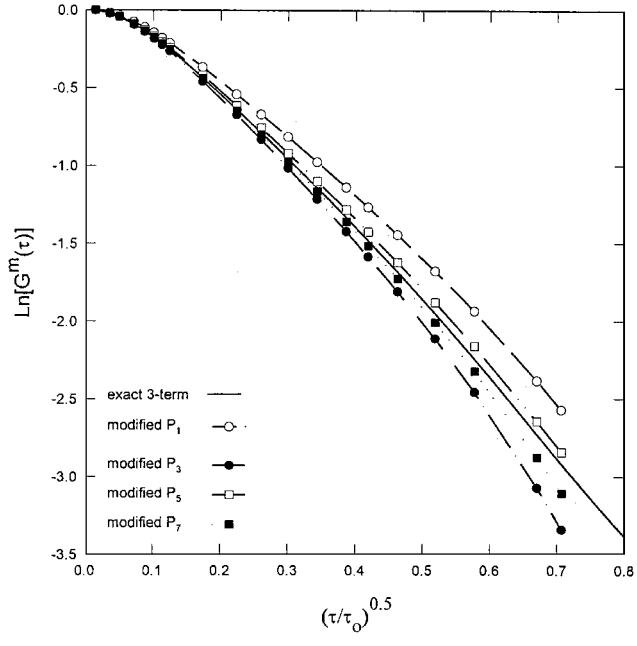

Fig. 2 Backscattering: Comparison of results P_N results with exact three-term Legendre expansion of g^1 for $L_0 = 5.0$.

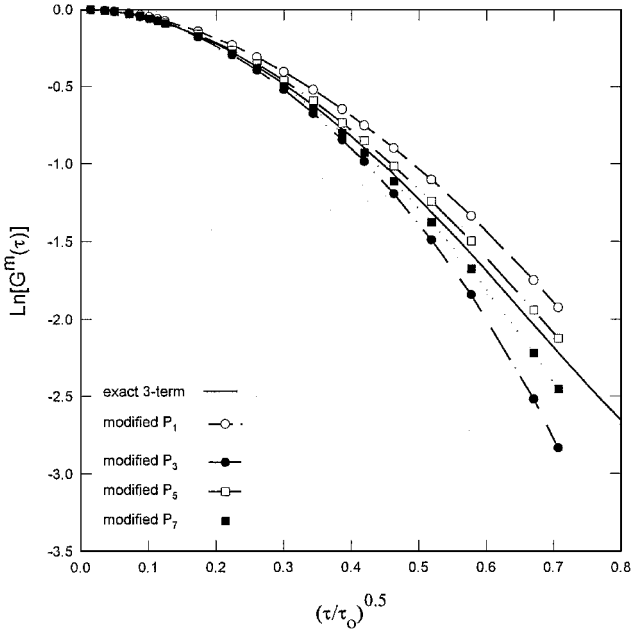

Fig. 1 Backscattering: Comparison of modified P_N results with exact three-term Legendre expansion of g^1 for $L_0 = 1.0$.

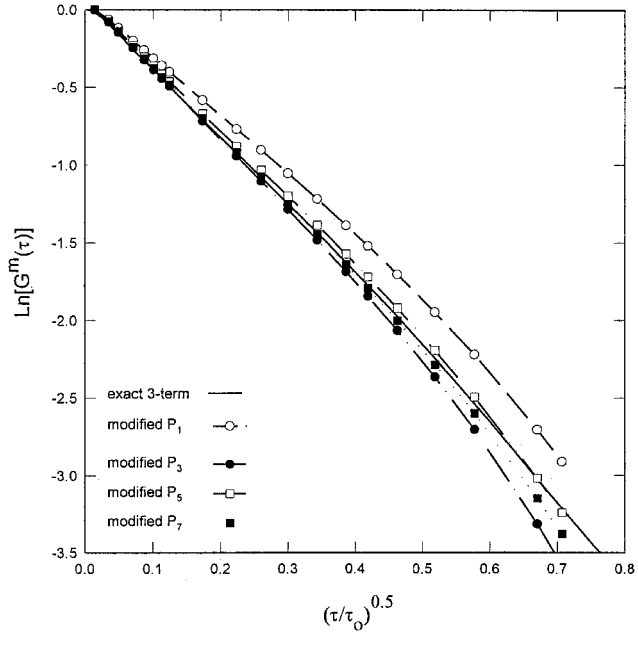

Fig. 3 Backscattering: Comparison of modified P_N results with exact three-term Legendre expansion of g^1 for $L_0 = 25.0$.

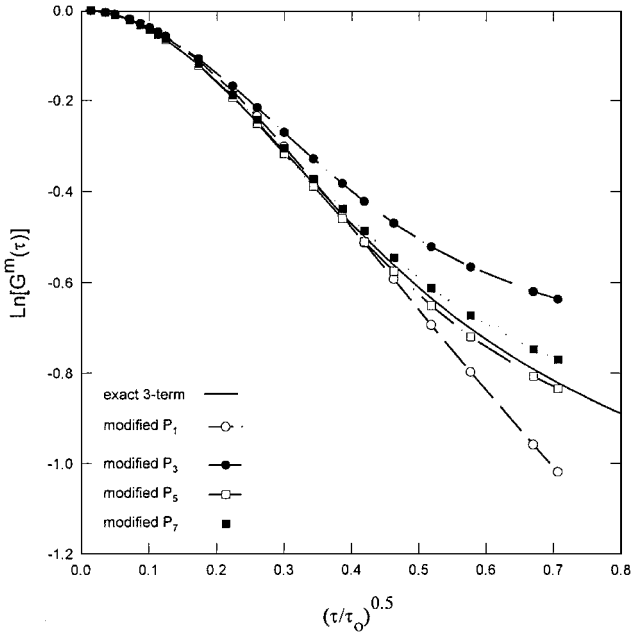

Fig. 4 Transmission: Comparison of modified P_N results with exact three-term Legendre expansion of g^1 for $L_0 = 1.0$.

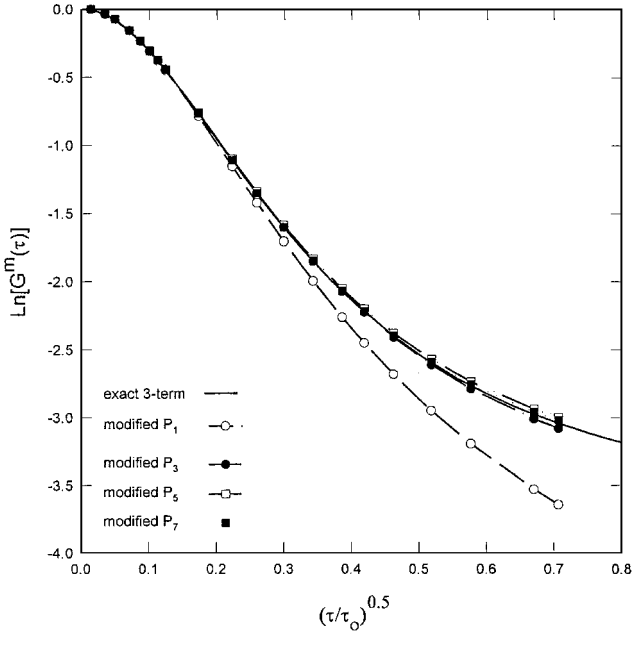

Fig. 5 Transmission: Comparison of modified P_N results with exact three-term Legendre expansion of g^1 for $L_0 = 5.0$.

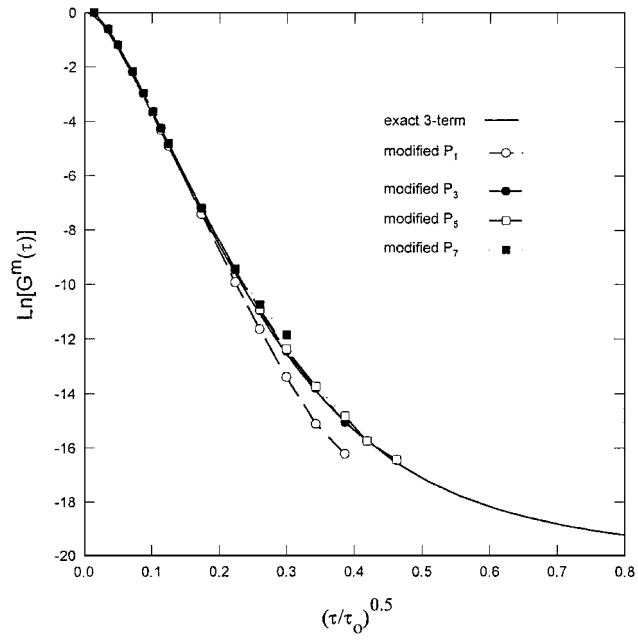

Fig. 6 Transmission: Comparison of modified P_N results with exact three-term Legendre expansion of g^1 for $L_0 = 25.0$.

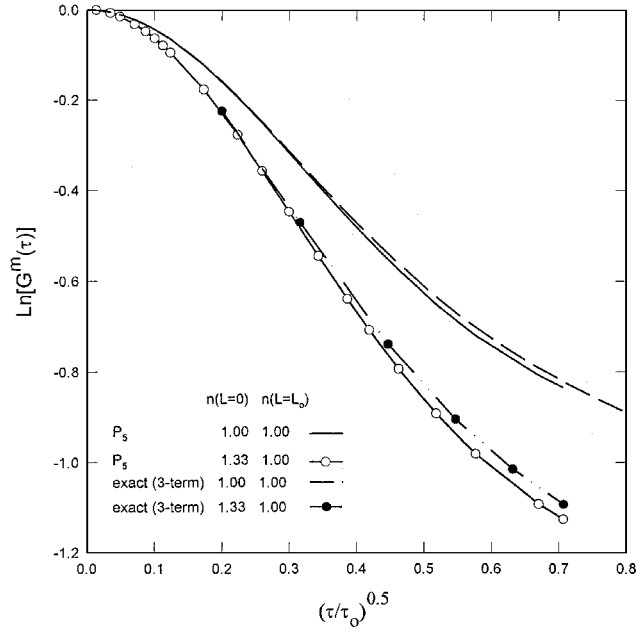

Fig. 7 Transmission: Comparison of modified P_5 results with exact three-term Legendre expansion of g^1 for $L_0 = 1.0$ with zero and one index of refraction change at the boundaries.

thicknesses of 1, 5, and 25. Note that the plots were normalized by the correlation at a delay time of $\tau/\tau_0 = 0.0002$. This is because the improved P_N approximation is not easily calculated at zero delay time due to a singularity problem. However, there is little difference in the solution of the CT equation for $\tau/\tau_0 = 0.0$ and 0.0002. Reguigui[37] showed good comparison between numerical results of the CT equation with a three-term g^1 expansion in a Legendre series for very small optical thickness to the actual single scattering g^1 correlation function for both transmission and backscattering up to a τ/τ_0 of about 0.6. He also found no significant improvement when the g^1 term in the CT equation was expanded up to eight terms.

Figures 1–3 show that the agreement between the modified P_N solution for backscattered correlation function and the three-term g^1 expansion solution, that is, exact three-term results, is improved for higher orders of P_N. The P_N results deviate from exact results for larger values of time due to the Legendre function behavior of the P_N approximation for the CT equation, but the two types of solutions agree well until a τ/τ_0 of about 0.5. The P_N solution alternatively over- and underestimates and converges to the exact three-term results as N increases. The agreement also improves as optical thickness increases because the medium becomes more diffuse, and the correlation function expansion in terms of Legendre polynomials will be more accurate for a smaller number of terms. (The P_1 approximation is the well-known diffusion approximation for very thick samples.) Because all P_N solutions begin curving down at longer time as compared to the exact three-term solution, at some point in time, it appears that the P_5 is better than the P_7 solution. In essence, all P_N solutions that lie above the exact results will eventually cross the exact results, appearing to be best at the crossing point. Thus, short time ($\tau/\tau_0 \leq 0.4$) solutions are better to examine for determining quality of solution. Figures 4–6 present very good comparisons for the third- and higher-order approximations to

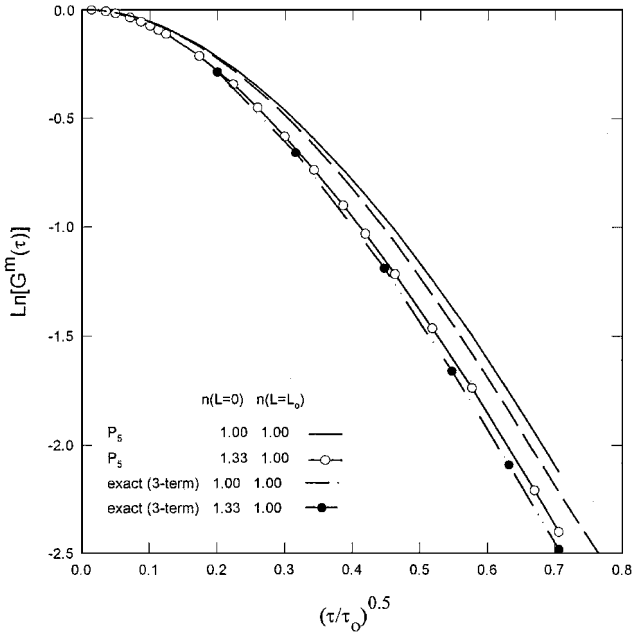

Fig. 8 Backscattering: Comparison of modified P_5 results with exact three-term Legendre expansion of g^1 for $L_0 = 1.0$ with zero and one index of refraction change at the boundaries.

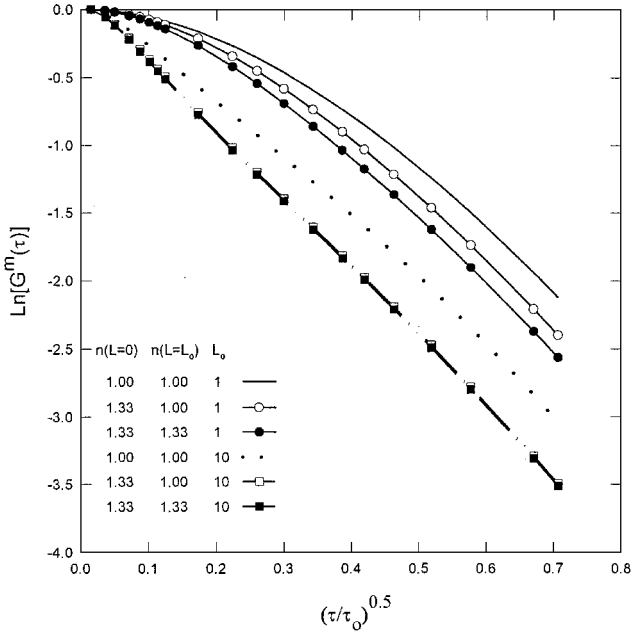

Fig. 9 Backscattering: Effect of refractive index change at the boundaries from modified P_5 results for $L_0 = 1.0$ and 10.

the exact three-term results for transmission at optical thicknesses greater than five. Note that the y axes for Figs. 4–6 do not have the same scale. The exact three-term results start curving up for larger delay time due to the g^1 Legendre expansion, which has a greater impact in transmission than backscattering. The correlation function decays much faster as optical thickness increases for transmission than for backscattering.

Figures 7 and 8 present the comparison of the correlation transfer results for the P_5 approximation to that of the exact three-term solution for transmission and backscattering at a small optical thickness of one when index of refraction at the $L = 0$ boundary changes. As shown, there is a significant effect on correlation function (especially in transmission) when index of refraction changes, and the P_5 results follow the same trend as the exact three-term results. The index of refraction change at the boundaries cause more scattering in the medium, and such a case appears to be optically thicker than the

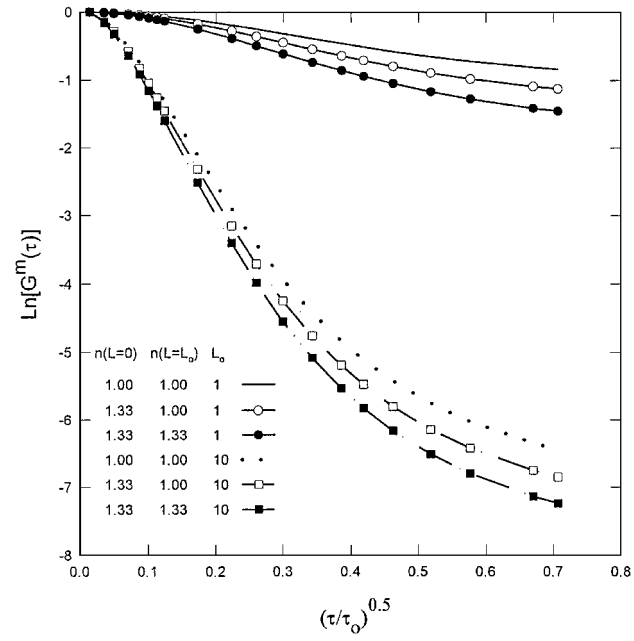

Fig. 10 Transmission: Effect of refractive index change at the boundaries from modified P_5 results for $L_0 = 1.0$ and 10.

case of unit refractive index, so that the correlation function decays faster.[29]

The effect of index of refraction has been shown in Figs. 9 and 10. Figure 9 presents modified P_5 results for the small and large optical thicknesses of 1 and 10 for backscattering. This shows the significant effect of index of refraction on the correlation function, and demonstrates that the $L = L_0$ boundary's index of refraction does not affect the results for high optical thickness. The portion of backscattering reflected from the $L = L_0$ boundary due to the nonunity index of refraction decays so much faster for high optical thickness that the signals corresponding to such scattering events have a negligible effect on the total backscattered correlation function. However, Fig. 10 shows that considering index of refraction changes at both boundaries is important at both small and large optical thicknesses for transmission.

The execution time for the modified P_N solution is comparable to that of standard P_N for the same order of N, but modified P_N accuracy is much better.[6,7] In addition, modified P_N computational time is much faster than that for the exact solution, more than 120 times faster (using a 133-MHz personal computer) for no index of refraction change at the boundary. The computational speed comparison increases much more (to more than 240 times) when considering index of refraction changes at the boundaries.

Conclusions

The CT equation, which is mathematically similar to the RT equation, can be used to provide important information on characteristics of a fluid and scattering particles suspended in the fluid. However, much computational time is required to obtain accurate, exact results, especially when considering such modeling parameters as index of refraction change through boundaries and interfaces. A modification of the classical spherical harmonics P_N method, which is one of the approximate techniques used to solve the radiative transfer equation, is presented herein to give a simple mathematical calculation for any order of N. For CT, a good comparison of the P_N results with the exact solution is presented for a higher order ($N > 3$), from optical thicknesses of 1–25. It has been shown that considering refractive index change across boundaries (or interfaces) affects both backscattering and transmission correlation results. Reduced execution time and good comparison between the exact solution and the modified P_N solution show that the P_N approximation can be successfully applied for correlation transfer problems if higher orders of N are employed.

Acknowledgments

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