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Self-consistent solutions to the equation of transfer with elastic and inelastic scattering in oceanic optics: I. Model

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A new self-consistent two-stream method has been developed that allows for both elastic and inelastic processes including fluorescence. What makes this method very useful is that it contains adjustable parameters that can be selected to fit experimental data. It also has the robustness to cover a complete range of inherent oceanic parameters ranging from the very clear to the most turbid. The method also uses real solar spectral input so that one can also perform chromaticity coordinate calculations for ocean color. Apparent optical properties such as irradiance and scalar irradiance can be computed at any depth in the ocean.

1. Introduction

Although the literature abounds with two-stream approximations[1–4] (these references contain a rather complete listing of most of the relevant papers), we present here an extension of a new two-stream method[1] that will allow for inelastic processes such as Raman scattering and fluorescence. What sets this method apart from all other two-stream approximations is the fact that it has the option of actually adjusting certain parameters (to be introduced later) to fit some experimental data obtained from either real marine waters such as the Mediterranean sea, Black sea, Atlantic ocean,[5] and Indian ocean[6] or emulated scattering and absorbing media.[5] This method also has the versatility to cover a complete range of inherent oceanic parameters ranging from the very clear to the very turbid. It allows for both molecular and hydrosol scattering as well as both chlorophyll and yellow-substance absorption. At present the model only allows for a homogeneous ocean with a flat surface; however, the extension to an inhomogeneous ocean with a stochastic interface can be obtained with relatively little work but will increase substantially the time per calculation. The input into this model can be quite arbitrary and allows for a realistic solar spectral input, which is a *sine qua non* for inelastic scattering processes.

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2. Basic Equations

A rigorous mathematical treatment of inelastic processes such as Raman scattering and fluorescence in hydrologic optics has not been presented in a systematic way in the open literature. For that reason we need to explain here the physics of our approach, which is based on the information given in Refs. 7, 8, and 9, and make the necessary relevant modifications to the equation of transfer. We have chosen here the widely accepted phenomenological approach,[10,11,12] which is less rigorous than the approach based on electrodynamics[13] but is reliable enough.

In the classic case (which neglects Raman scattering and fluorescence) the light energy is absorbed and scattered elastically. The absorbed part of the energy, which is proportional to the absorption coefficient a, was regarded as totally converted to thermal energy; however, in reality some of the absorbed energy reappears in fluorescence. The scattered part of the energy, which is proportional to the scattering coefficient b, is not lost but reemerges as elastically scattered light at the same wavelength. There is no transfer of light energy from one wavelength to another.

When we include inelastic scattering processes a new problem arises because the process of the light energy transfer starts to depend on the light distribution and optical properties of the medium at a number of different wavelengths. Fortunately, quite similar problems have already been treated many years ago in neutron transport theory and have come to be known as multispeed transport.[11] The modification of this method to our problem is quite simple; it

implies that the total scattering coefficient b consists of both the Raman part b^R and the elastic part b^E and adds the corresponding source term to the source part of the equation of transfer. The principal difference between Raman and elastic source terms is the presence of summation (or integration) over excitation wavelengths in the Raman term.

According to the latest studies,[8,14] it is more productive to regard fluorescence in sea water, not as a scattering process but as a process of re-emission of light energy by the substances of biological origin such as chlorophyll a and dissolved organic matter (DOM); this re-emission peaks at fixed wavelengths (approximately 685 nm for the chlorophyll a or red fluorescence and near 425 nm for the DOM or blue fluorescence). The lifetimes for these fluorescent processes are typically of the order of nanoseconds, which is sufficient time for the emitted photon to lose memory of the direction of the incident photon, which also accounts for the isotropy of the fluorescent radiation phase function. Because we do not consider fluorescence as a scattering process, we do not make any fluorescent correction to the scattering coefficient. The only modification regarding the effect of fluorescence is the addition of a source function term. The physics of the fluorescent process adopted in this study is totally consistent with the approach used previously in the research of Gordon.[9]

One of the most frequently used equations for consistency checking of numerical results is Gershun's equation.[15] We provide a brief derivation of this equation when inelastic effects are present. We start with the scalar transfer equation for spectral radiance $L(\lambda, \mathbf{r}, \Omega)$[4,10,11] in a medium that allows inelastic effects such as Raman scattering and fluorescence:

$$[\mathbf{n}\nabla + c(\lambda)]L(\lambda, \mathbf{r}, \Omega) = \frac{b^E(\lambda)}{4\pi}\int_\Omega p(\cos\Theta)L(\lambda, \mathbf{r}, \Omega')\mathrm{d}\Omega' + \frac{1}{4\pi}\sum_{I=R,F}\int_{\lambda'<\lambda}\mathrm{d}\lambda'\sigma^I(\lambda', \lambda) \times \int_\Omega p^I(\cos\Theta)L(\lambda', \mathbf{r}, \Omega')\mathrm{d}\Omega', \quad (1)$$

where $\mathbf{n}$ is a vector in the direction of propagation of light, $c = a + b^E + b^R$ is the extinction coefficient, b^E is an elastic scattering coefficient by density fluctuations (Rayleigh) and hydrosol particles, b^R is a Raman-scattering coefficient, $a = a^{th} + a^{Fl}$ is an absorption coefficient, which consists of a thermal dissipation part a^{th} and a fluorescent absorption part a^{Fl}, $\Theta = \cos^{-1}[\cos\theta\cos\theta' + \sin\theta\sin\theta'\cos(\varphi - \varphi')]$ is a scattering angle, θ, φ are the zenith and azimuthal angles of the radiation, respectively, $p(\cos\Theta)$ is an elastic scattering phase function, and $p^I(\cos\Theta)$ are inelastic angular emission functions; all of them (p and p^I) are normalized according to the equation

$$\int_\Omega p(\cos\Theta)\mathrm{d}\Omega \equiv \int_0^{2\pi}\mathrm{d}\varphi\int_0^{\pi}p(\cos\Theta)\sin\theta\,\mathrm{d}\theta = 4\pi, \quad (2)$$

where $\mathrm{d}\Omega = \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi$ is an element of solid angle, λ is the wavelength of light, $\mathbf{r}$ is a position coordinate vector, $\sigma^I(\lambda', \lambda)$, ($I = R, F$) are differential emission coefficients for Raman scattering (index R) and fluorescence (index F), which are connected with total inelastic emission coefficients ϵ^I by the equation

$$\epsilon^I(\lambda) = \frac{1}{4\pi}\int_{\lambda'<\lambda}\sigma^I(\lambda', \lambda)\mathrm{d}\lambda'\int_\Omega p^I(\cos\Theta)\mathrm{d}\Omega', \quad I = R, F. \quad (3)$$

These terms allow for energy transport from shorter wavelengths into the observation wavelength. It should also be noted that the Raman-scattering coefficient $b^R(\lambda)$ and the fluorescent part of the absorption coefficient $a^{Fl}(\lambda)$ are also connected with differential emission coefficients through the following equations:

$$b^R(\lambda) = \frac{1}{4\pi}\int_{\lambda'>\lambda}\sigma^R(\lambda, \lambda')\mathrm{d}\lambda'\int_\Omega p^R(\cos\Theta)\mathrm{d}\Omega', \quad (4)$$

$$a^{Fl}(\lambda) = \frac{1}{4\pi}\sum_{F=C,Y}\int_{\lambda'>\lambda}\mathrm{d}\lambda'\sigma^F(\lambda, \lambda')\int_\Omega p^F(\cos\Theta)\mathrm{d}\Omega', \quad (5)$$

where indices C and Y, respectively, denote red fluorescence by chlorophyll and blue fluorescence by yellow substance. These terms allow for energy transport out of the observation wavelength region λ to longer wavelengths.

Introducing scalar and vector irradiances according to

$$E_0 = \int_\Omega L(\Omega)\mathrm{d}\Omega, \quad (6)$$

$$\mathbf{E} = \int_\Omega L(\Omega)\mathbf{n}\mathrm{d}\Omega, \quad (7)$$

and integrating Eq. (1) over solid angle, we get the analog of Gershun's equation for the inelastic case, namely,

$$\mathrm{div}\,\mathbf{E} = -[a^{th} + \Delta a]E_0, \quad (8)$$

where $\Delta a = (a^{Fl} - \epsilon^{Fl}) + (b^R - \epsilon^R)$, $\epsilon^{Fl} = \epsilon^C + \epsilon^Y$. Equations for a^{Fl}, ϵ^{Fl}, b^R, and ϵ^R are given in Appendix A. A heuristic derivation of Eq. (8) for the case of Raman scattering was given previously in the research of Stavn and Weidemann.[16]

Because we are only interested in the laterally

homogeneous case we will rewrite Eq. (1) for the spectral radiance $L(\lambda, z, \theta, \varphi)$ in the form

$$\left[\cos\theta \frac{d}{dz} + c(\lambda)\right] L(\lambda, z, \theta, \varphi) = Q(\lambda, z, \theta, \varphi), \quad (9)$$

$$Q(\lambda, z, \theta, \varphi) = Q^E(\lambda, z, \theta, \varphi) + \sum_{I=R,C,Y} Q^I(\lambda, z, \theta, \varphi) + Q^{HO}(\lambda, z, \theta, \varphi), \quad (10)$$

where $Q^E(\lambda, z, \theta, \varphi)$ is the elastic scattering source function, $\Sigma_{I=R,C,Y} Q^I(\lambda, z, \theta, \varphi)$ is a sum of the inelastic source terms, $Q^{HO}(\lambda, z, \theta, \varphi)$, which will be neglected below, describes higher orders of inelastic scattering, i.e., higher than first order (Raman scattering and fluorescence or any combination of them), z is a vertical coordinate (Oz has its origin on the surface and is considered positive in the downward direction), and θ and φ are zenith and azimuthal angles between the Oz axis and the direction of propagation, so that $0 \le \theta \le \pi/2$ gives downward radiation and $\pi/2 < \theta \le \pi$ gives upward radiation. We represent the total radiance as

$$L(\lambda, z, \theta, \varphi) = L^E(\lambda, z, \theta, \varphi) + \sum_{I=R,C,Y} L^I(\lambda, z, \theta, \varphi); \quad (11)$$

here $L^E(\lambda, z, \theta, \varphi)$ is the elastic radiance, i.e., radiance in the absence of Raman and fluorescent effects, and $L^I(\lambda, z, \theta, \varphi)$ is the inelastic radiance, $I = R$ (Raman) or F (fluorescence), $F = C$ (red fluorescence) or $F = Y$ (blue fluorescence).

The source terms can be represented as follows:

$$Q^E(\lambda, z, \theta, \varphi) = \frac{b^E(\lambda)}{4\pi} \int p(\cos\Theta) L^E(\lambda, z, \theta, \varphi) d\Omega', \quad (12)$$

$$Q^R(\lambda, z, \theta, \varphi) = \frac{b^E(\lambda)}{4\pi} \int p(\cos\Theta) L^R(\lambda, z, \theta', \varphi') d\Omega' + \frac{1}{4\pi} \int_{\lambda' < \lambda} d\lambda' \sigma^R(\lambda', \lambda) \times \int p^R(\cos\Theta) L^E(\lambda', z, \theta', \varphi') d\Omega', \quad (13)$$

$$Q^F(\lambda, z, \theta, \varphi) = \frac{b^E(\lambda)}{4\pi} \int p(\cos\Theta) L^F(\lambda, z, \theta', \varphi') d\Omega' + \frac{1}{4\pi} \int_{\lambda' < \lambda} d\lambda' \sigma^F(\lambda', \lambda) \times \int p^F(\cos\Theta) L^E(\lambda', z, \theta', \varphi') d\Omega', \quad F = C, Y, \quad (14)$$

$$\sigma^R(\lambda', \lambda) = \frac{d\tilde{\nu}'}{d\lambda'} \sigma_\nu^R(\tilde{\nu}', \tilde{\nu}) = \frac{\tilde{\nu}'^2}{k_\nu} \beta_0^R \left(\frac{\tilde{\nu}'}{\tilde{\nu}_0}\right)^4 f^R(\tilde{\nu}', \tilde{\nu}); \quad (15)$$

$$\sigma^F(\lambda', \lambda) = \beta_0^F a_0^F(\lambda') C_F f^F(\lambda', \lambda), \quad F = C, Y, \quad (16)$$

where $\tilde{\nu}' = k_\nu/\lambda'$, and $\tilde{\nu} = k_\nu/\lambda$ (in inverse centimeters) are excitation and emission wave numbers, respectively, $k_\nu = 1 = 10^7$ nm/cm, $\tilde{\nu}_0 = k_\nu/400$ nm = 25,000 cm^{-1}, a_0^F is a specific absorption coefficient of the fluorescent substance, C_F is a concentration of that substance, β_0^F is its fluorescence efficiency, and β_0^R is the Raman-scattering coefficient at $\lambda = 400$ nm; $f^R(\tilde{\nu}', \tilde{\nu})$ (in centimeters) is the Raman excitation function, normalized according to the following equation:

$$\int_{-\infty}^{+\infty} f^R(\tilde{\nu}', \tilde{\nu}) d\tilde{\nu}' = \int_{-\infty}^{+\infty} f^R(\tilde{\nu}', \tilde{\nu}) d\tilde{\nu} = 1; \quad (17)$$

the fluorescence distribution function $f^F(\lambda', \lambda)$ (in inverse nanometers squared) is represented as

$$f^F(\lambda', \lambda) = f_{ex}^F(\lambda') f_{em}^F(\lambda), \qquad \int_{-\infty}^{+\infty} f_{ex}^F(\lambda) d\lambda = 1,$$

$$\int_{-\infty}^{+\infty} f_{em}^F(\lambda) d\lambda = 1, \qquad F = C, Y, \quad (18)$$

where f_{ex}^F and f_{em}^F excitation and emission functions, respectively (see Appendix A), and $p^R(\cos\Theta)$ is the Raman phase function given by[17]

$$p^R(\cos\Theta) = \frac{1 + 3k_\rho \cos^2\Theta}{1 + k_\rho}, \qquad k_\rho \approx 0.2, \quad (19)$$

$p^F(\cos\Theta)$ is the fluorescence angular emittance function, which is taken to be isotropic,[8] so $p^F(\cos\Theta) = 1$ sr^{-1}, $F = C, Y$ (for details concerning Raman and fluorescence models see Appendix A).

Introducing the operator

$$\hat{T}\varphi \equiv \left[\cos\theta \frac{d}{dz} + c - \frac{b^E}{4\pi} \int d\Omega' p(\cos\Theta)\right] \varphi, \quad (20)$$

we can divide Eq. (9) for total radiance L [Eq. (11)] into two separate equations for elastic and inelastic radiances which will be solved separately in Sections 3 and 4 of this paper

$$\hat{T} L^E(\lambda, z, \theta, \varphi) = 0, \quad (21)$$

$$\hat{T} L^I(\lambda, z, \theta, \varphi) = \frac{1}{4\pi} \int_{\lambda' < \lambda} d\lambda' \int d\Omega' \sigma^I(\lambda', \lambda) p^I(\cos\Theta) \times L^E(\lambda', z, \theta', \varphi'), \quad I = R, F, \quad F = C, Y. \quad (22)$$

3. Approximate Solutions for the Elastic Anisotropically Scattering Part

In this section we will obtain a solution to Eq. (21) for the irradiances that will be as simple as possible and yet have a precision of 5–10% and still be valid for the

complete range of variability of inherent optical properties, i.e., from very clear to very turbid ocean waters. In the absence of Raman scattering and fluorescence the exact equation for the scalar radiance is

$$\left[\cos\theta\frac{\mathrm{d}}{\mathrm{d}z}+c\right]L^E(z,\theta,\varphi)=\frac{b^E}{4\pi}\int p(\cos\Theta)L^E(z,\theta',\varphi')\mathrm{d}\Omega', \quad (23)$$

where we have dropped the wavelength variable λ because we are only considering elastic scattering.

In this section we will use the approach found in Khalturin[1]; namely, we represent the highly anisotropic sea water volume scattering function in the form $p(\cos\Theta)=p_T(\cos\Theta)+p_\Delta(\cos\Theta)$, where $p_T(\cos\Theta)=2B+2(1-2B)\delta(1-\cos\Theta)$ is the transport phase function (see Davison,[11] p. 241, and also Refs. 18 and 19), $B=0.5\int_{\pi/2}^{\pi}p(\cos\theta)\sin\theta\mathrm{d}\theta$ is the backscattering probability, $\delta(x)$ is the Dirac delta function, and $p_\Delta=p-p_T$. We will retain only the transport part, p_T, of the phase function and take into account the influence of p_Δ indirectly by adjusting mutual dependencies of mean cosines of the scattered light with empirical relationships derived from experimental results. In order not to overload this paper with the details and discussions already published we will use only what is logically necessary from Ref. 1 plus the necessary modifications for inelastic effects.

Let us introduce the renormalized[18,19] optical depth $\tau=\epsilon z$, where $\epsilon=\tilde{a}+2b_B$ is the renormalized attenuation coefficient, $b_B=b^EB$ is the elastic backscattering coefficient, $\tilde{a}=c-b^E\equiv a+b^R$, and $\tilde{x}=b_B/(\tilde{a}+b_B)$. This renormalization is necessary because the approach of Ref. 1 implies that the predominant part of the forward-scattered rays are excluded from the scattered light, which leads to the renormalization $b\to 2b_B$. Additional renormalization $a\to a+b^R$ is due to Raman scattering. The peculiarity of this renormalization is the fact that Raman scattering at a given wavelength effectively increases extinction simply because all Raman energy is Stokes shifted to longer wavelengths. The fluorescence (at least in the interpretation adopted here: $a=a^{th}+a^{Fl}$) does not affect any inherent optical property because it originated from re-emittance of energy already taken from the light by ordinary absorption (coefficient a). Using these parameters in Eq. (23), we get

$$\left(\mu\frac{\mathrm{d}}{\mathrm{d}\tau}+1\right)L^E(\tau,\mu,\varphi)=\frac{1}{2\pi}\left[\frac{\tilde{x}}{1+\tilde{x}}\int_0^{2\pi}\mathrm{d}\varphi'\int_{-1}^{1}\mathrm{d}\mu' L^E(\tau,\mu',\varphi')+\Delta(\tau,\mu,\varphi)\right], \quad (24)$$

where

$$\Delta(\tau,\mu,\varphi)=\frac{\tilde{\omega}_0(1-\tilde{x})}{2(1-\tilde{\omega}_0)(1+\tilde{x})}\int_0^{2\pi}\mathrm{d}\varphi'\int_{-1}^{1}\mathrm{d}\mu'\times[p(\mu')-p_T(\mu')]L^E(\tau,\mu',\varphi'), \quad (25)$$

where $\mu=\cos\theta$, $\mu'=\cos\theta'$, $\tilde{\omega}_0=b^E/(\tilde{a}+b^E)=b^E/c$ is the single-scattering albedo.

Let us represent the elastic radiance as a sum of *scattered* and *unscattered* light[20]

$$L^E=L^s+L^q. \quad (26)$$

The *unscattered* radiance L^q should satisfy the equation

$$\left(\mu\frac{\mathrm{d}}{\mathrm{d}\tau}+1\right)L^q(\tau,\mu,\varphi)=0, \quad (27)$$

with the boundary condition $L^q(0,\mu,\varphi)=L_0^q(\mu,\varphi)$, $\mu>0$. It is necessary to note here that because L^q is a sum of unscattered light and forward-scattered rays, Eq. (27) is not obvious. It is a direct consequence of our approach and can be used only if the method used in Refs. 1 and 18 is applicable, which is always the case for sea water.

The solution of Eq. (27) will be

$$L^q(\tau,\mu,\varphi)=L_0^q(\mu,\varphi)\exp(-\tau/\mu). \quad (28)$$

Then *scattered* radiance should satisfy the equation

$$\hat{T}_\Delta L^s=\frac{g(\tau)}{2\pi}, \quad (29)$$

where

$$\hat{T}_\Delta L^s\equiv\left(\mu\frac{\mathrm{d}}{\mathrm{d}\tau}+1\right)L^s(\tau,\mu,\varphi)-\frac{1}{2\pi}\left[\frac{\tilde{x}}{1+\tilde{x}}\int_0^{2\pi}\mathrm{d}\varphi'\times\int_{-1}^{1}\mathrm{d}\mu' L^s(\tau,\mu',\varphi')+\Delta(\tau,\mu,\varphi)\right], \quad (30)$$

with the boundary conditions

$$L^s(0,\mu,\varphi)|_{\mu>0}=0, \qquad \lim_{\tau\to\infty}L^s(\tau,\mu,\varphi)=0, \quad (31)$$

where

$$g(\tau)=\frac{\tilde{x}}{1+\tilde{x}}\int_0^{2\pi}\mathrm{d}\varphi\int_{-1}^{1}\mathrm{d}\mu L_0^q(\mu,\varphi)\exp(-\tau/\mu). \quad (32)$$

It should be noted that the function $\Delta(\tau,\mu,\varphi)$ vanishes in two cases: isotropic scattering ($B=1/2$) and complete forward scattering ($B=0$).

To rewrite Eq. (29) in terms of irradiances, we must introduce the following definitions[21]:

$$E_d^s = \int_0^{2\pi} d\varphi \int_0^1 L^s(\mu, \varphi)\mu d\mu,$$

$$E_u^s = -\int_0^{2\pi} d\varphi \int_{-1}^0 L^s(\mu, \varphi)\mu d\mu, \tag{33}$$

$$E_{0d}^s = \int_0^{2\pi} d\varphi \int_0^1 L^s(\mu, \varphi) d\mu,$$

$$E_{0u}^s = \int_0^{2\pi} d\varphi \int_{-1}^0 L^s(\mu, \varphi) d\mu, \tag{34}$$

$$E_0^s = \int_0^{2\pi} d\varphi \int_{-1}^1 L^s(\mu, \varphi) d\mu = E_{0d}^s + E_{0u}^s. \tag{35}$$

Let us now introduce the following mean cosines[21]:

$$\bar{\mu}_d = \frac{E_d^s}{E_{0d}^s}, \qquad \bar{\mu}_u = \frac{E_u^s}{E_{0u}^s},$$

$$\bar{\mu} = \frac{E_d^s - E_u^s}{E_0^s} = \frac{E_d^s - E_u^s}{E_{0d}^s + E_{0u}^s}. \tag{36}$$

Applying the operators $\int_0^{2\pi} d\varphi \int_0^1 d\mu \ldots$ and $\int_0^{2\pi} d\varphi \int_{-1}^0 d\mu \ldots$ to Eq. (29), which neglects $\Delta(\tau, \mu, \varphi)$, and then taking Δ into account indirectly by using the empirical relations[2,5,6] $\bar{\mu}_d = 1/(2 - \bar{\mu})$ and $\bar{\mu}_u = 1/(2 + \bar{\mu})$, we get the following system of equations:

$$\sum_{\beta=1,2} \hat{L}_{\alpha\beta}(\tau) E_\beta^s = e_\alpha g(\tau),$$

$$\hat{\mathbf{L}}(\tau) = \begin{bmatrix} \frac{d}{d\tau} + q_- & -\tilde{x}q_+ \\ -\tilde{x}q_- & -\frac{d}{d\tau} + q_+ \end{bmatrix}, \tag{37}$$

where

$$\mathbf{e} = \begin{bmatrix} 1 \\ 1 \end{bmatrix},$$

$$q_\pm = \frac{2 \pm \bar{\mu}}{1 + \tilde{x}}.$$

Any Greek index here assumes values: d, u, or 1, 2, $(1 \leftrightarrow d, 2 \leftrightarrow u)$.

We should note here that any two-flow approximation reduces the transfer equation to a system of differential equations for the downward and upward irradiances. This system is dependent on two unknown parameters, namely, $\bar{\mu}_d$, and $\bar{\mu}_u$. Different two-flow approximations adopt different values for $\bar{\mu}_d$ and $\bar{\mu}_u$, usually without any regard to experimental data. In this paper we have adopted the experimental empirical equations $(\bar{\mu}_\alpha = 1/[2 + (-1)^\alpha \bar{\mu}]$, $\alpha = 1, 2)$, which connect $\bar{\mu}_d$ and $\bar{\mu}_u$ $(\bar{\mu}_1 \equiv \bar{\mu}_d, \bar{\mu}_2 \equiv \bar{\mu}_u)$ with the mean cosine $\bar{\mu}$, which can be calculated from the exact result (see Ref. 1):

$$k_\infty = \frac{\tilde{a}}{\epsilon\bar{\mu}} \equiv \frac{1 - \tilde{x}}{\bar{\mu}(1 + \tilde{x})}. \tag{38}$$

The system of Eqs. (37) has two eigenvalues, $-k_\infty$ and k_0. Let us impose the last part of the self-consistency principle,[1] which consists of the following two steps. The first is substituting the transport[11,18,19] phase function $p_T(\cos\Theta)$ for the arbitrary highly anisotropic phase function $p(\cos\Theta)$, which drastically simplifies the equation to be solved at the expense of precision (see discussion of this method in Refs. 1 and 2). The second is reclaiming the main part of the lost precision by (a) using experimental[5,6] empirical relations between total, upward, and downward mean cosines and (b) equating the values given by the exact equation for the eigenvalue given by Eq. (38) with the corresponding eigenvalue of Eqs. (37), which is

$$k_\infty = \frac{\bar{\mu} - [4 - (4 - \bar{\mu}^2)\tilde{x}^2]^{1/2}}{(1 + \tilde{x})}. \tag{39}$$

This method[22] has some shortcomings, but in our case it works well if we restrict ourselves to the integral optical properties and irradiances. Using the results above, we get the following equations for $\bar{\mu}$, k_∞, and k_0:

$$\bar{\mu} = \left\{\frac{1 + 2\tilde{x} - [\tilde{x}(4 + 5\tilde{x})]^{1/2}}{1 + \tilde{x}}\right\}^{1/2}$$

$$\equiv \left\{\frac{1 - \tilde{x}}{1 + 2\tilde{x} + [\tilde{x}(4 + 5\tilde{x})]^{1/2}}\right\}^{1/2}, \quad \tilde{x} = \frac{(1 - \bar{\mu}^2)^2}{1 + 4\bar{\mu}^2 - \bar{\mu}^4}, \tag{40}$$

$$k_\infty = \frac{\bar{\mu}(3 - \bar{\mu}^2)}{1 + \bar{\mu}^2}, \qquad k_0 = \bar{\mu}(4 - \bar{\mu}^2). \tag{41}$$

For simplicity we restrict ourselves to the case of illumination of the sea surface by direct sunlight. In this case

$$L_0^q(\mu, \varphi) = L_0\delta(\varphi)\delta(\mu - \mu_s), \quad \mu_s = \left(1 - \frac{\sin^2 z_\odot}{n_r^2}\right)^{1/2}, \tag{42}$$

where $z_\odot$ is the solar zenith angle and n_r is the refractive index of sea water. The source function $g(\tau)$ has the form

$$g(\tau) = \frac{\tilde{x}L_0}{1 + \tilde{x}} \exp(-\tau/\mu_s), \tag{43}$$

and the solution to Eqs. (37) with the boundary conditions

$$E_d^s(z=0)=0, \qquad \lim_{z\to\infty} E_i^s(z)=0 \tag{44}$$

for an optically infinitely deep ocean will be

$$\begin{cases} E_d^s(\tau)=A[\exp(-k_\infty\tau)-\exp(-\tau/\mu_s)] \\ E_u^s(\tau)=R_\infty A\exp(-k_\infty\tau)-B\exp(-\tau/\mu_s), \end{cases} \tag{45}$$

where

$$R_\infty=\left(\frac{1-\bar{\mu}}{1+\bar{\mu}}\right)^2 \tag{46}$$

is the diffuse reflectance in the asymptotic regime (where the radiance distribution is independent of solar zenith direction), and

$$A=\frac{\tilde{x}L_0(2+\bar{\mu}+1/\mu_s)}{(1+\tilde{x})(1/\mu_s-k_\infty)(1/\mu_s+k_0)}, \qquad B=\frac{\tilde{x}L_0(2-\bar{\mu}-1/\mu_s)}{(1+\tilde{x})(1/\mu_s-k_\infty)(1/\mu_s+k_0)}, \tag{47}$$

or

$$\begin{cases} E_d^s(\tau)=L_0\mu_s R_s\dfrac{Q_s m_1}{1-\mu_s k_\infty} \\ \qquad\qquad \times[\exp(-k_\infty\tau)-\exp(-\tau/\mu_s)] \\ E_u^s(\tau)=L_0\mu_s R_s\left\{\exp(-k_\infty\tau)+\dfrac{Q_s m_2}{1-\mu_s k_\infty}\right. \\ \qquad\qquad \left.\times[\exp(-k_\infty\tau)-\exp(-\tau/\mu_s)]\right\}, \end{cases} \tag{48}$$

where

$$m_1=1+\mu_s(2+\bar{\mu}), \qquad m_2=\mu_s(2-\bar{\mu})-1,$$

$$Q_s=\frac{1}{1+R_\infty}\equiv\frac{(1+\bar{\mu})^2}{2(1+\bar{\mu}^2)},$$

$$R_s=\frac{(1-\bar{\mu})^2}{1+\mu_s k_0}\equiv\frac{(1-\bar{\mu})^2}{1+\mu_s\bar{\mu}(4-\bar{\mu}^2)}, \tag{49}$$

where R_s is the diffuse reflectance of the sea illuminated by direct sunlight. *Unscattered* irradiances are

$$E_d^q(\tau)=L_0\mu_s\exp(-\tau/\mu_s), \qquad E_u^q(\tau)\equiv 0. \tag{50}$$

We can rewrite the total elastic irradiances $E_\alpha^E=E_\alpha^q+E_\alpha^s$ and their derivatives in a form convenient for numerical computation:

$$E_d^E(z)=L_0\mu_s\exp(-\nu z)[\exp(2z_s)+R_s\eta Q_s m_1 D_0(z)], \tag{51}$$

$$E_u^E(z)=L_0\mu_s R_s\exp(-\nu z)[1+\eta Q_s m_2 D_0(z)], \tag{52}$$

$$-\frac{dE_d^E(z)}{dz}=L_0\mu_s\exp(-\nu z)\eta[\exp(2z_s)+R_s Q_s m_1 D_z(z)], \tag{53}$$

$$-\frac{dE_u^E(z)}{dz}=L_0\mu_s R_s\exp(-\nu z)[\nu+\eta Q_s m_2 D_z(z)], \tag{54}$$

where $\eta=\epsilon/\mu_s$, $\nu=\epsilon k_\infty$, $z_s=(\nu-\eta)z/2$, $D_0(z)=(z/2)[1+\exp(2z_s)]f_\tau(z_s)$, $f_\tau(x)=\tanh(x)/x$, $D_z(z)=\nu D_0(z)-\exp(2z_s)$.

Downward and upward elastic irradiance attenuation coefficients will be

$$k_d^E(z)=-\frac{d\ln E_d^E(z)}{dz}=\eta\frac{\exp(2z_s)+R_s Q_s m_1 D_z(z)}{\exp(2z_s)+R_s\eta Q_s m_1 D_0(z)}, \tag{55}$$

$$k_u^E(z)=-\frac{d\ln E_u^E(z)}{dz}=\frac{\nu+\eta Q_s m_2 D_z(z)}{1+\eta Q_s m_2 D_0(z)}. \tag{56}$$

4. Approximate Solutions for Inelastic Scattering (Raman Scattering and Fluorescence)

Equation (22) in explicit form is

$$\left(\mu\frac{d}{d\tau}+1\right)L^I(\tau,\mu,\varphi)=\frac{1}{2\pi}\left[\frac{\tilde{x}}{1+\tilde{x}}\int_0^{2\pi}d\varphi'\int_{-1}^{1}d\mu' L^I(\tau,\mu',\varphi')+\Delta(\tau,\mu,\varphi)+g^{sI}(\tau,\mu,\varphi)+g^{qI}(\tau,\mu,\varphi)\right], \tag{57}$$

or

$$\hat{T}_\Delta L^I=Q^I, \qquad Q^I=\frac{1}{2\pi}(g^{sI}+g^{qI}), \qquad I=R,F, \qquad F=C,Y, \tag{58}$$

where

$$g^{sR}(\lambda,\tau,\mu,\varphi)=\frac{3}{8\epsilon(\lambda)}\int_{\lambda'<\lambda}d\lambda'\sigma^R(\lambda',\lambda)\int_0^{2\pi}d\varphi'\int_{-1}^{1}d\mu'\times\frac{1+3k_\rho\cos^2\Theta}{1+k_\rho}L^s(\lambda',\tau,\mu',\varphi'), \tag{59}$$

$$g^{sF}(\lambda,\tau,\mu,\varphi)=\frac{1}{2\epsilon(\lambda)}\int_{\lambda'<\lambda}d\lambda'\sigma^F(\lambda',\lambda)\int_0^{2\pi}d\varphi'\times\int_{-1}^{1}d\mu' L_0^s(\lambda',\mu',\varphi'), \tag{60}$$

$$g^{qR}(\lambda,\tau,\mu,\varphi)=\frac{3}{8\epsilon(\lambda)}\int_{\lambda'<\lambda}d\lambda'\sigma^{R}(\lambda',\lambda)\int_0^{2\pi}d\varphi'\int_0^1 d\mu'\times\frac{1+3k_p\cos^2\Theta}{1+k_p}L_0^q(\lambda',\mu',\varphi')\times\exp\left[-\frac{\tau(\lambda')}{\mu'}\right],\quad(61)$$

$$g^{qF}(\lambda,\tau,\mu,\varphi)=\frac{1}{2\epsilon(\lambda)}\int_{\lambda'<\lambda}d\lambda'\sigma^{F}(\lambda',\lambda)\int_0^{2\pi}d\varphi'\int_0^1 d\mu'\times L_0^q(\lambda',\mu',\varphi')\exp\left[-\frac{\tau(\lambda')}{\mu'}\right],\quad(62)$$

and

$$\cos\Theta=\mu\mu'+(1-\mu^2)^{1/2}(1-\mu'^2)^{1/2}\cos(\varphi-\varphi').\quad(63)$$

Eqs. (58) differ from Eq. (29) only by right-hand sides. They can be reduced to equations for irradiances in a similar manner. We now have

$$\sum_{\beta=1,2}\hat{L}_{\alpha\beta}(\tau)E_\beta^I(\tau)=e_\alpha g^I(\tau),$$

$$\mathbf{e}=\begin{bmatrix}1\\1\end{bmatrix},\qquad \alpha,\beta=d,u\text{ or }1,2,\quad(64)$$

with the boundary conditions

$$E_d^I(z=0)=0,\qquad \lim_{z\to\infty}E_\alpha^I(z)=0,\qquad \alpha=d,u.$$

Here the matrix operator $\hat{\mathbf{L}}(\tau)$ has the form of Eqs. (37) and the right-hand sides $g^I(\tau)$ of Eqs. (64) become

$$g^I(\lambda,z)=\frac{1}{2\epsilon(\lambda)}\int_{\lambda'<\lambda}d\lambda'\sigma^I(\lambda',\lambda)\left\{L_0(\lambda')\exp\left[-\frac{\epsilon(\lambda')z}{\mu_s}\right]+[2-\bar{\mu}(\lambda')]E_d^s(\lambda',z)+[2+\bar{\mu}(\lambda')]E_u^s(\lambda',z)\right\},\quad(65)$$

where the irradiances E_d^s and E_u^s are given by Eqs. (48).

The solution to Eqs. (64) can be represented as

$$\mathbf{E}^I(\tau)=A^I\mathbf{a}\exp(-k_\infty\tau)+\int_0^\infty\tilde{\mathbf{G}}(\tau-\tau')g^I(\tau')d\tau',\quad(66)$$

with

$$\mathbf{E}^I=\begin{bmatrix}E_1^I\\E_2^I\end{bmatrix},\qquad \mathbf{a}=\begin{bmatrix}1\\R_\infty\end{bmatrix},\qquad \tilde{\mathbf{G}}(\tau)=\begin{bmatrix}G_{11}(\tau)+G_{12}(\tau)\\G_{21}(\tau)+G_{22}(\tau)\end{bmatrix},$$

where the constant A^I is determined from the boundary conditions and $\mathbf{G}(\tau)$ is the Green's function matrix,[23] which satisfies the equation

$$\sum_{\beta=1,2}\hat{L}_{\alpha\beta}(\tau)G_{\beta\gamma}(\tau)=\delta_{\alpha\gamma}\delta(\tau),\quad(67)$$

where $\delta_{\alpha\beta}$ is the Kronecker delta. It is not difficult to show,[1] that

$$\mathbf{G}(\tau)=\begin{bmatrix}1 & R_0\\ R_\infty & R_0R_\infty\end{bmatrix}\frac{\theta(\tau)\exp(-k_\infty\tau)}{1-R_0R_\infty}+\begin{bmatrix}R_0R_\infty & R_0\\ R_\infty & 1\end{bmatrix}\frac{\theta(-\tau)\exp(k_0\tau)}{1-R_0R_\infty},\quad(68)$$

and

$$\tilde{\mathbf{G}}(\tau)=k_1[\mathbf{a}k_2\theta(\tau)\exp(-k_\infty\tau)+\mathbf{b}\theta(-\tau)\exp(k_0\tau)],\quad(69)$$

where

$$R_0=R_\infty\frac{2+\bar{\mu}}{2-\bar{\mu}},\qquad \mathbf{b}=\begin{bmatrix}R_0\\1\end{bmatrix},$$

$$k_1=\frac{1+R_\infty}{1-R_0R_\infty},\qquad k_2=\frac{1+R_0}{1+R_\infty},$$

where $\theta(x)$ is the Heavyside function. Solutions to Eqs. (64) with the boundary condition $E_d^I(0)=0$ that vanish at infinite depths are

$$\mathbf{E}^I(\tau)=k_1\left(\mathbf{a}\left\{k_2\int_0^\tau g^I(\tau')\exp[-k_\infty(\tau-\tau')]d\tau'-R_0\exp(-k_\infty\tau)\int_0^\infty g^I(\tau')\exp(-k_0\tau')d\tau'\right\}+\mathbf{b}\int_\tau^\infty g^I(\tau')\exp[k_0(\tau-\tau')]d\tau'\right).\quad(70)$$

Equation (70) together with Eqs. (51) and (52) for elastic irradiances gives the solution to our problem. Unfortunately Eq. (70) contains many factors in the expression for $g^I(\tau)$ that become indeterminate at some wavelengths and unsuitable for numerical calculations without further analytical modification. This task can be easily performed,[24] and the results are

$$\mathbf{E}^I(\lambda,z)=\frac{k_1}{2}\exp(-\nu z)\langle\mathbf{a}(k_2D_R-R_0L_R)+\mathbf{b}L_N\rangle^I,\quad(71)$$

$$-\frac{d\mathbf{E}^I(\lambda,z)}{dz}=\frac{k_1}{2}\exp(-\nu z)\{\langle\nu[\mathbf{a}(k_2D_R-R_0L_R)+\mathbf{b}L_N]\rangle^I-\langle(\nu+\zeta)\mathbf{b}L_N\rangle^I+\langle(\mathbf{b}-k_2\mathbf{a})[(1+kD_3)\times\exp(2r_1)+n\exp(2r_2)]\rangle^I\},\quad(72)$$

where

$$l_1 = 1/(\zeta + \eta'), \quad \zeta = \epsilon k_0, \quad l_2 = 1/(\zeta + \nu'),$$

$$r_1 = (\nu - \eta')z/2, \quad r_2 = (\nu - \nu')z/2, \quad r_3 = r_2 - r_1,$$

$$D_i = (z/2)[1 + \exp(2r_i)]f_\tau(r_i), \quad i = 1, 2, 3,$$

$$D_R = D_1 + nD_2 + kS, \quad n = \mu_s R_s'(2 + \bar{\mu}'),$$

$$S = (z^2/4)\{[\exp(2r_1) + \exp(2r_2)]f_\tau(r_1)f_\tau(r_3) - [1 + \exp(2r_2)]f_{xy}(r_1, r_2)\},$$

$$k = 2\mu_s R_s' Q_s' \eta'[\mu_s(4 - \bar{\mu}'^2) - \bar{\mu}'],$$

$$L_R = l_1 + l_2(n + kl_1), \quad L_1 = l_1 \exp(2r_1),$$

$$L_N = L_1 + l_2 N_K + L_K, \quad N_K = (n + kl_1)\exp(2r_2),$$

$$L_K = kD_3 L_1, \qquad f_{xy}(x, y) = \frac{1}{y - x}\left[\frac{\tanh(x)}{x} - \frac{\tanh(y)}{y}\right].$$

Any primed parameter here denotes dependence on primed wavelength, i.e., $\varsigma \equiv \varsigma(\lambda)$, $\varsigma' \equiv \varsigma(\lambda')$. The angular brackets $\langle \Psi(\lambda', \lambda)\rangle^I$ denote integration over wavelength according to the rule $\langle \Psi(\lambda', \lambda)\rangle^I \equiv \int d\lambda' \sigma^I(\lambda', \lambda) L_0(\lambda') \Psi(\lambda', \lambda)$, where $\sigma^I(\lambda', \lambda)$ are given by Eqs. (15) and (16) and $\Psi(\lambda', \lambda)$ can be any function.

The total irradiance attenuation coefficient[12,25] can be calculated according to

$$k_\alpha = -\frac{d}{dz}\ln\left[E_\alpha^E(z) + \sum_{I=R,C,Y} E_\alpha^I(z)\right] = k_\alpha^E + \frac{\sum_{I=R,C,Y}(k_\alpha^I - k_\alpha^E)\rho_\alpha^I}{1 + \sum_{I=R,C,Y}\rho_\alpha^I}, \qquad \alpha = d, u, \quad (73)$$

where $\rho_\alpha^I = E_\alpha^I / E_\alpha^E$, or

$$\rho_d^I = \frac{k_1\langle k_2 D_R - R_0 L_R + R_0 L_N\rangle^I}{2L_0\mu_s[\exp(2z_s) + R_s\eta Q_s m_1 D_0]},$$

$$\rho_u^I = \frac{k_1\langle R_\infty(k_2 D_R - R_0 L_R) + L_N\rangle^I}{2L_0\mu_s R_s[1 + \eta Q_s m_2 D_0]}, \quad (74)$$

elastic irradiance attenuation coefficients k_d^E and k_u^E are given, correspondingly, by Eqs. (55) and (56), and coefficients k_d^I and k_u^I are given by the following equations:

$$\begin{cases} k_d^I = -\dfrac{d}{dz}\ln E_d^I(z) = \nu + \dfrac{\langle (R_0 - k_2)[(1 + kD_3)\exp(2r_1) + n\exp(2r_2)] - (\nu + \zeta)R_0 L_N\rangle^I}{\langle k_2 D_R - R_0 L_R + R_0 L_N\rangle^I} \\ k_u^I = -\dfrac{d}{dz}\ln E_u^I(z) = \nu + \dfrac{\langle (1 - k_2 R_\infty)[(1 + kD_3)\exp(2r_1) + n\exp(2r_2)] - (\nu + \zeta)L_N\rangle^I}{\langle R_\infty(k_2 D_R - R_0 L_R) + L_N\rangle^I}. \end{cases} \quad (75)$$

The total diffuse reflectance will be

$$R \equiv \frac{E_u(z = 0)}{E_d(z = 0)} = R_s + \sum_{I=R,C,Y}\delta R^I, \quad (76)$$

where

$$\delta R^I = \frac{1 + R_\infty}{2L_0\mu_s}\langle l_1 + l_2(n + kl_1)\rangle^I. \quad (77)$$

5. Accuracy of the Method

A. Accuracy With Purely Elastic Scattering

We shall evaluate the accuracy of some of the formulas for elastic scattering by comparing them with the results obtained by a number of other authors. We shall compare the values we have obtained using our formulas with the approximate results of other authors,[26] using quite accurate numerical calculations[27,28] and experimental data.[29,30]

The relative error of the asymptotic regime attenuation coefficient

$$\gamma = \frac{1 - \omega_0}{\bar{\mu}}, \quad \text{where } \bar{\mu} = \left\{\frac{1 + 2x - [x(4 + 5x)]^{1/2}}{1 + x}\right\}^{1/2}, \quad (78)$$

is shown in Fig. 1; here

$$x = \frac{b_B}{a + b_B} \equiv \frac{\omega_0 B}{1 - \omega_0(1 - B)}.$$

The exact values of γ computed in Ref. 28 for different values of ω_0 and B were compared with the values computed with our approximate Eqs. (78). In the range of typical marine waters, i.e., single-scattering albedos $\omega_0 \le 0.6$ and backscattering probabilities $B \le 0.17$ errors incurred by using Eqs. (78) are less than 6%.

Figure 2 illustrates the behavior of R/x (R is a diffuse reflectance) as a function of Gordon's parameter x. We compared our data calculated with the approximate Eq. (46), shown by the stars, with the experimental data of Timofeyeva[29] (shown by the solid squares), exact numerical Monte-Carlo represen-

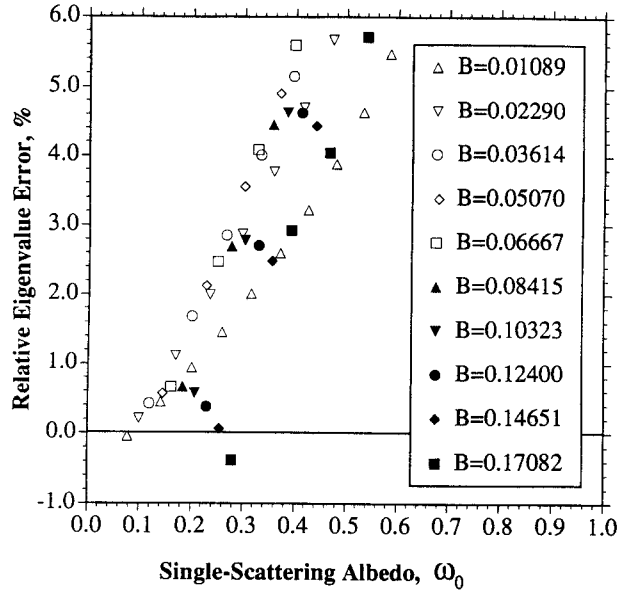

Fig. 1. Relative error of the calculation of the eigenvalue [Eqs. (78)] as a function of single-scattering albedo ω_0 for different values of backscattering probability B. Exact values are taken from Loskutov.[28]

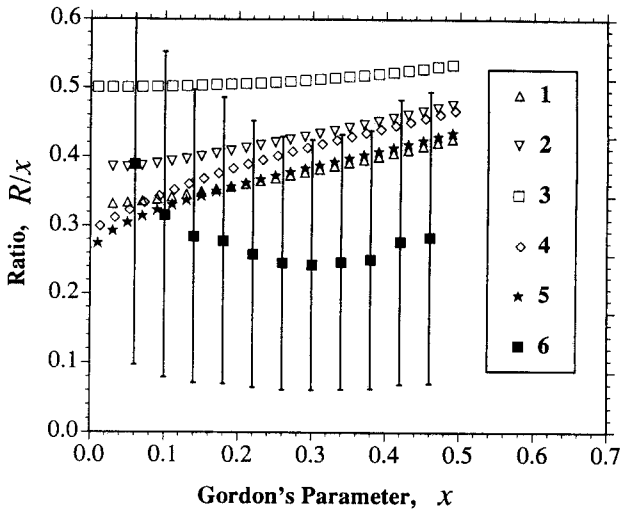

Fig. 2. Ratio of diffuse reflectance R to Gordon's parameter x as a function of x according to different sources: (1) Monte-Carlo calculations for direct solar illumination from the nadir by Gordon *et al.*,[27] (2) Monte-Carlo calculations for diffuse illumination by Gordon *et al.*,[27] (3) Kubelka-Munk,[26] (4) exact values for a special type of scattering phase function,[31] (5) our method [see Eq. (46)], (6) experimental data by Timofeyeva.[29]

tations by Gordon *et al.*,[27] namely,

$$R_G^{dir} = 0.0001 + 0.3244x + 0.1425x^2 + 0.1308x^3, \tag{79}$$

for direct solar illumination (open upward triangles), and for diffuse illumination (open downward triangles):

$$R_G^{diff} = 0.0003 + 0.3687x + 0.1802x^2 + 0.0740x^3. \tag{80}$$

We also used the Kubelka–Munk formula[26] (open squares), namely,

$$R_{KM} = \frac{[1 - (1 - x^2)^{1/2}]}{x}. \tag{81}$$

Finally we compared our results with values calculated with the exact formula for a special type of scattering phase function[31] (open diamonds):

$$R_H = \frac{1 - \chi}{1 + \chi}[(1 + \chi^2)^{1/2} - \chi]^2,$$

$$\text{where } \chi = \left[\frac{1 - x}{1 + (3 + 2\sqrt{2})x}\right]^{1/2}. \tag{82}$$

It is easily seen that Eq. (46) gives the closest agreement with the exact and experimental values in comparison with the approximate broadly accepted Eq. (81).

Additional estimates given in Refs. 1 and 2 show that this approach gives estimates for elastic integral optical characteristics in the range of 15% precision for $0 \le x \le 1$ and $\omega_0 \le 0.6$.

B. Accuracy With a Combination of Elastic and Inelastic Scattering

To estimate the actual accuracy of our equations for irradiances, namely, Eqs. (51) and (52) for E_d^E and E_u^E, respectively, and Eq. (71) for E_d^R and E_u^R, we compared our results with the Raman-scattering Monte-Carlo calculations of Kattawar and Xu[17] with the same inherent optical properties. For the Monte-Carlo calculations we processed one million histories at the excitation wavelength to get the inelastic component, and then we processed another one million histories to get the elastic component. These two calculations took roughly 4 hr on a Silicon Graphics 4D/340S computer (36 Megaflops). For the present model, however, calculations on the Macintosh IIcx (0.4 Megaflops), which produced results for 165 wavelengths and 51 depths, lasted only 8 min. The time of calculation with the present model, if executed on the same type of computer, is roughly 10^5 times less than the corresponding (for all 165 wavelengths) Monte-Carlo calculation. The inherent op-

Table 1. Optical Properties Used for the Monte-Carlo Simulation and Analytical Calculations Displayed in Figs. 3(a) and 3(b)[a]

λ (nm)	a (m^{-1})	b^{Hyd} (m^{-1})	b^{Water} (m^{-1})
417	0.03289	0.0443	0.00487
440	0.03007	0.0416	0.00386
486	0.02669	0.0373	0.00251
518	0.04894	0.0354	0.00191

[a]For Raman scattering the corresponding excitation → emission wavelengths are 417 → 486 nm and 440 → 518 nm.

ing to the data given in Refs. 9 and 38, namely,

$$\sigma^C(\lambda', \lambda) = \beta_0{}^C a_0{}^C(\lambda') C_C f^C(\lambda', \lambda), \tag{A4}$$

$$f^C(\lambda', \lambda) = f_{ex}{}^C(\lambda') f_{em}{}^C(\lambda),$$

$$f_{em}{}^C(\lambda) = k^C \exp\left[-\frac{(\lambda - \lambda^{C0})^2}{2\sigma_C{}^2}\right], \tag{A5}$$

where $\beta_0{}^C = 0.0289$ nm, $a_0{}^C$ is the specific absorption coefficient of chlorophyll in square meters per milligram, C_C is a chlorophyll concentration in milligrams per cubic meter, $\lambda^{C0} = 685$ nm, $\sigma_C = 10.6$ mm, and $k^C = 0.037636$ nm^{-1}.

The wavelength redistribution of light emitted due to fluorescence by DOM or yellow substance[35]:

$$\sigma_v{}^Y(\lambda', \lambda) = \beta_0{}^Y a_0{}^Y(\lambda') C_Y f^Y(\lambda', \lambda), \tag{A6}$$

$$f^Y(\lambda', \lambda) = f_{ex}{}^Y(\lambda') f_{em}{}^Y(\lambda),$$

$$f_{em}{}^Y(\lambda) = k^Y \exp\left[-\frac{(\lambda - \lambda^{Y0})^2}{2\sigma_Y{}^2}\right], \tag{A7}$$

where $\beta_0{}^Y = 0.92$ nm, $a_0{}^Y$ is a specific absorption coefficient by yellow substance in inverse meters, C_Y is a concentration of yellow substance (dimensionless), $\lambda^{Y0} = 425$ nm, $\sigma_Y = 50$ nm, and $k^Y = 0.00665$ nm^{-1}. The chlorophyll excitation function can be represented as[9]

$$f_{ex}{}^C(\lambda) = \begin{cases} h_{ex}{}^C(\lambda/\lambda^{C0}), & \lambda_1{}^C \le \lambda \le \lambda_2{}^C \\ 0, & \text{elsewhere} \end{cases}, \tag{A8}$$

where $h_{ex}{}^C = 5.06 \cdot 10^{-4}$ cm, $\lambda_1{}^C = 370$ nm, $\lambda_2{}^C = 690$ nm. We do not have enough information concerning the shape of the $f_{ex}{}^Y(\lambda)$, but in general it behaves similar to $f_{ex}{}^C(\lambda)$ with different parameters; i.e., it is negligible at wavelengths outside the interval between $\lambda_1{}^Y = 250$ nm and $\lambda_2{}^Y = 400$ nm and may have a different coefficient before λ inside this interval. Quantum efficiency η^C for chlorophyll fluorescence is taken to be $\eta^C = \beta_0{}^C h_{ex}{}^C = 0.008$ ($\equiv 0.8\%$).[9]

The fluorescence parts of the absorption coefficient and fluorescence emittance are represented by

$$a^{Fl}(\lambda) = \beta_0{}^C \langle a_0{}^C \rangle_{em} C_C f_{ex}{}^C(\lambda) + \beta_0{}^Y \langle a_0{}^Y \rangle_{em} C_Y f_{ex}{}^Y(\lambda), \tag{A9}$$

$$\epsilon^{Fl}(\lambda) = \beta_0{}^C \langle a_0{}^C \rangle_{ex} C_C f_{em}{}^C(\lambda) + \beta_0{}^Y \langle a_0{}^Y \rangle_{ex} C_Y f_{em}{}^Y(\lambda), \tag{A10}$$

where

$$\langle a_0{}^F \rangle_{ex} = \int_{-\infty}^{\infty} a_0{}^F(\lambda) f_{ex}{}^F(\lambda) d\lambda,$$

$$\langle a_0{}^F \rangle_{em} = \int_{-\infty}^{+\infty} a_0{}^F(\lambda) f_{em}{}^F(\lambda) d\lambda, \qquad F = C, Y.$$

The angular distribution of Raman-scattered light is given by Eq. (19), and the angular distribution of light emitted that is due to fluorescence is isotropic.

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