

Light-scattering coefficient of seawater for arbitrary concentrations of hydrosols

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The scattering coefficient of water as a function of concentration of hydrosol particles is calculated. A new quantum-mechanical approach to calculate the multiple-scattering phenomenon in seawater is proposed. The approach is based on Maxwell's equations for the light fields in stochastically scattering water with hydrosols. The water is modeled as a thermally fluctuating medium filled with the particles. It is found that at small concentrations of scatterers the scattering coefficient is linear in the concentration. At higher values of concentrations the dependence on the concentration may be approximated by a power law. © 1999 Optical Society of America [S0740-3232(99)01007-8]

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1. INTRODUCTION

In a majority of publications available today, the light-scattering coefficient of water is considered linearly dependent on the concentration of hydrosols.¹ *In situ* experiments⁴ show that at certain concentrations of chlorophyll C_C , typical of coastal waters, the dependence of the water absorption coefficient on C_C is nonlinear. It is known⁵ that the scattering coefficient of water is nonlinearly dependent on chlorophyll concentration, which is strongly correlated to the concentration of scattering matter of biologic origin.

In this paper an attempt is made to develop a new approach to calculate the scattering coefficient of water as a function of concentration of hydrosol particles C_p . The approach is based on the solutions of Maxwell's equations in a stochastically scattering medium⁶ (water). The water is modeled as a thermally fluctuating medium filled with the hydrosol scatterers.

The result of this paper is Eq. (63) in Section 6 for the water scattering coefficient. The scattering coefficient b is linear in the concentration of scattering particles at values typical of the open ocean. The coefficient depends quadratically on the concentration when the concentration is very small (typical of the Sargasso Sea waters). At concentrations close to the values that are typical of coastal waters, the dependence on concentration weakens and reaches saturation at very high values. The results of this paper can explain some experimental data obtained in turbid coastal waters.

To investigate this problem, we start from Maxwell's equations in a stochastically scattering and absorbing medium. The mathematical formalism of scattering in a stochastic medium is identical to the formalism of quantum statistical mechanics.^{6,7} The photons remain discrete quantum particles.⁸⁻¹⁰ The principal challenges are the mathematics involved in these calculations and the fidelity of the modeling of the physics of photon scattering. Fortunately, the computational techniques have already been developed and tested.^{6,8,11} Because the

quantum-mechanical approach in ocean optics is new, let us show that this approach is relevant to the physics of the light-scattering phenomenon in water.

As a starting point that will help to rationalize the motivation for this new approach, let us estimate the density of solar photons near the sea surface. According to Ref. 12, the irradiance by sunlight near the sea surface, L_\otimes , is less than $1.353 \times 10^3 \text{ W/m}^2$. At the same time the energy flux that is due to a single photon at 500-nm wavelength is

$$s_{\text{ph}} = \hbar \bar{\omega} c \approx 10^{-10} \text{ W m}$$

$$\left(\bar{\omega} = \frac{2\pi c}{n_w \bar{\lambda}}, \quad \bar{\lambda} = 500 \text{ nm} \equiv 5 \times 10^{-7} \text{ m} \right), \quad (1)$$

where \hbar is the Planck constant, c is the speed of light in vacuum, $\bar{\omega}$ and $\bar{\lambda}$ are, respectively, the average visible light frequency and wavelength, and $n_w \approx 1.34$ is the water refractive index. Consequently, the density of the solar photons near the sea surface, ρ_{ph}^\otimes , is

$$\rho_{\text{ph}}^\otimes = \frac{L_\otimes}{\hbar \bar{\omega} c} \leq 10^{13} \text{ m}^{-3} \approx (50 \text{ } \mu\text{m})^{-3}. \quad (2)$$

The estimate given by Eq. (2) shows that one photon near the sea surface at one moment of time occupies a volume that is larger than a cube with 50- μm sides. According to Ref. 13 the total concentration of particles with the radii range 0.01–1 μm is approximately 5×10^{12} – 10^{13} m^{-3} . It means that one photon is scattering on one or more particles. This is clearly a situation for quantum-mechanical consideration. The classical approach implies that the number of photons should be large enough to neglect quantum phenomena.

The question arises as to why the classical theory is so good for ocean optics. The answer is simple: This is a result of the process of measurements. Let us consider the process of measurement. Let a photoreceiver with the time constant τ_r record the radiance falling on the

sensor area S_r . In this case the photoreceiver is averaging the signal from the ensemble of photons that are enclosed in the volume

$$V_r = S_r \tau_r c = N_{\text{ph}} V_{\text{ph}}. \quad (3)$$

The number N_{ph} of photons averaged by the photoreceiver is

$$N_{\text{ph}} = \frac{S_r \tau_r L_{\infty}}{\hbar \bar{\omega}} = L_{\infty} \frac{S_r \tau_r \bar{\lambda} n_w}{2 \pi \hbar c}. \quad (4)$$

As a typical photoreceiver let us choose the device with $\tau_r = 0.02$ s and $S_r = 0.05$ cm². For this device the averaging volume is $V_r = 30$ m³. This volume corresponds to averaging of 10^{14} – 10^{15} photons near the sea surface or $N_{\text{ph}} \sim (10^{14} - 10^{15}) T_a$ below the sea surface (here T_a is an attenuation factor). For typical hydro-optical devices N_{ph} is always large enough to make the measurement results look classical.

2. APPROACH

The radiative transfer theory, predominantly used in ocean optics,^{14–18} cannot explain the nonlinear dependence of the inherent optical properties on the concentration of scatterers. The correct derivation of the scalar radiative transfer equation^{18–23} goes through the following chain: Maxwell's equations → Dyson's and Bethe–Salpeter's equations → vector radiative transfer equation → scalar radiative transfer equation.²⁴ Each step in this chain is connected with a loss of precision and a loss of opportunity to derive certain physical properties. At the level of the vector radiative transfer equation, the opportunity to derive inherent optical properties as a function of concentrations of dissolved and suspended matter and other physical properties of the medium is already lost.

In this paper the scattering coefficient of hydrosol particles is calculated through the dielectric permittivity of the hydrosol component in the water. The dielectric permittivity is a constituent part of the Fourier transform of the Green's function of Maxwell's equations in water. By definition, the Green's function is a solution of these equations when a source function is assumed as infinitely short and localized at one point light pulse.²⁶

3. INTERACTION HAMILTONIAN

To solve a scattering problem in classical electrodynamics, we need a vector or scalar wave equation derived from Maxwell's equations. This equation includes a time-dependent and inhomogeneous-in-space term proportional to the dielectric permittivity of the medium. Different ways of averaging the electric fields, its binary products, and dielectric permittivity fields produce various theories of light propagation in a scattering medium. It is possible to reformulate the classical Maxwellian theory in terms of quantum mechanics. This approach is based on the concept of a scattering matrix that is defined as

$$S = \hat{T} \exp \left[-i \int_0^{\infty} \hat{H}_{\text{int}}(t) dt \right]. \quad (5)$$

Here \hat{H}_{int} is an interaction Hamiltonian, i.e., the change in the energy of the system after the introduction of scattering elements with subsequent replacement of the physical values (velocities, vector potentials, etc.) by corresponding operators; t is the time; and \hat{T} is the ordering operator: it rearranges the operators in Eq. (5) in such a manner that the operators taken at a later time always appear to the left of the operators taken at earlier times. Let us assume that the seawater with hydrosols has the wave functions $|0\rangle$ and $|\infty\rangle$ before and after the act of photon scattering. Then these functions are coupled with the following equation:

$$|0\rangle = S|\infty\rangle \equiv \hat{T} \exp \left[-i \int_0^{\infty} \hat{H}_{\text{int}}(t) dt \right] |\infty\rangle. \quad (6)$$

All averaging procedures in quantum-mechanical scattering involve the scattering matrix given by Eq. (5). Now we derive the interaction Hamiltonian between photons and absorbing and scattering seawater filled with hydrosol particles.

Let us use a standard method for the derivation of the interaction Hamiltonian.²⁷ The change in the energy E of water after insertion of hydrosol particles is

$$\delta E = (4\pi)^{-1} \int d^3r \mathbf{E}(\mathbf{r}, t) \delta \mathbf{D}(\mathbf{r}, t), \quad (7)$$

where $\mathbf{E}(\mathbf{r}, t)$ is the electric field, $\delta \mathbf{D}(\mathbf{r}, t)$ is the change in the electric displacement $\mathbf{D}(\mathbf{r}, t)$ related to the difference between the dielectric permittivity of hydrosol particles and the dielectric permittivity of surrounding water, \mathbf{r} is a spatial coordinate, and t is the time. The electric displacement inside a hydrosol particle is²⁸

$$\mathbf{D}_m(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}, t) + \int_0^{\infty} f_m(t') \mathbf{E}(\mathbf{r}, t - t') dt', \quad (8)$$

where m denotes the number of the particle and the function f_m determines the dispersion properties of the m th hydrosol particle. For further convenience let us introduce the following Heaviside-type²⁶ function:

$$U_m(\mathbf{r} - \mathbf{r}_m) = \begin{cases} 1, & \mathbf{r} \in V_m \\ 0, & \mathbf{r} \notin V_m \end{cases}, \quad (9)$$

which is equal to unity inside the m th particle and equal to zero outside (here \mathbf{r}_m is a vector that points to the gravity center of the m th particle, and V_m is the volume of the m th particle). Using the function U_m , we write down the change in the energy of the water caused by insertion of hydrosols. The change in electric displacement is

$$\delta \mathbf{D}(\mathbf{r}, t) = \int_0^{\infty} \delta f(\mathbf{r}, t') \mathbf{E}(\mathbf{r}, t - t') dt', \quad (10)$$

$$\delta f(\mathbf{r}, t') = \sum_m \langle U_m(\mathbf{r} - \mathbf{r}_m) \rangle_m [f_m(t') - f_w(t')], \quad (11)$$

where $f_w(t)$ is a dispersion function of the water without hydrosols. The angle brackets $\langle \cdot \rangle_m$ denote averaging

over all possible positions of \mathbf{r}_m . The summation is made over all hydrosol particles in the water volume V_0 .

According to Ref. 28, the distribution function of hydrosol particles can be represented as a series over the powers of water density:

$$\begin{aligned} F(\mathbf{r}, t) &\cong \alpha_1 \rho(\mathbf{r}, t) + \alpha_2 \rho^2(\mathbf{r}, t) + \dots, \\ \int F(\mathbf{r}, t) d^3r &= 1, \quad \rho(\mathbf{r}, t) = \rho_0 + \delta\rho(\mathbf{r}, t), \\ \rho_0 &\approx 1 \text{ g/cm}^3, \end{aligned} \quad (12)$$

where $\rho_0 = V_0^{-1} \int \rho(\mathbf{r}, t) d^3r$ is the average water density in the volume V_0 and $\delta\rho(\mathbf{r}, t)$ is the water density fluctuation. Let us keep only the first term in relation (12); then we have

$$F(\mathbf{r}, t) \cong \frac{\rho(\mathbf{r}, t)}{V_0 \rho_0} = \frac{1}{V_0} + \frac{\delta\rho(\mathbf{r}, t)}{V_0 \rho_0}, \quad (13)$$

$$\begin{aligned} \langle U_m(\mathbf{r} - \mathbf{r}_m) \rangle_m &= \int F(\mathbf{r}', t) U_m(\mathbf{r} - \mathbf{r}') d^3r' \\ &= C_m + \frac{1}{V_0 \rho_0} \int \delta\rho(\mathbf{r}, t) U_m(\mathbf{r} - \mathbf{r}') d^3r, \end{aligned} \quad (14)$$

where $C_m = V_m/V$. In this case

$$\begin{aligned} \delta f(\mathbf{r}, t') &= \sum_m C_m [f_m(t') - f_w(t')] \\ &+ \frac{1}{\rho_0} \int \delta\rho(\mathbf{r}', t') v(\mathbf{r} - \mathbf{r}', t') d^3r', \end{aligned} \quad (15)$$

where

$$v(\mathbf{r}, t) = \frac{1}{V_0} \sum_m U_m(\mathbf{r}) [f_m(t) - f_w(t)]. \quad (16)$$

For the changes in the electric displacement, we have

$$\delta \mathbf{D}(\mathbf{r}, t) = \delta \mathbf{D}_0(\mathbf{r}, t) + \delta \mathbf{D}_s(\mathbf{r}, t), \quad (17)$$

where

$$\delta \mathbf{D}_0(\mathbf{r}, t) = \sum_m C_m \int_0^\infty dt' [f_m(t') - f_w(t')] \mathbf{E}(\mathbf{r}, t - t'), \quad (18)$$

$$\begin{aligned} \delta \mathbf{D}_s(\mathbf{r}, t) &= \rho_0^{-1} \int_0^\infty dt' \int d^3r' \delta\rho(\mathbf{r}', t) \\ &\times v(\mathbf{r} - \mathbf{r}', t - t') \mathbf{E}(\mathbf{r}, t - t'). \end{aligned} \quad (19)$$

The first correction term in Eq. (17) is not related to the scattering. It determines a correction to the water dielectric permittivity that is due to the introduction of hydrosols. In Section 4 we incorporate this term into the original water dielectric permittivity by renormalizing its value.

When the volume concentration of hydrosol particles is small ($C_V \equiv \sum_m C_m \ll 10^{-3}$), it is possible to neglect dispersion properties of particles. In this case we have

$$f_m(t) - f_w(t) \cong (\epsilon_m - \epsilon_{0w}) \delta(t + 0), \quad (20)$$

where ϵ_m is the dielectric permittivity of the m th hydrosol particle, ϵ_{0w} is the dielectric permittivity of the water without particles, and $\delta(t)$ is the Dirac delta function.²⁶ Taking into account relation (20), we have

$$v(\mathbf{r}, t) = \delta\epsilon_h C_V \Delta(\mathbf{r}) \delta(t + 0), \quad (21)$$

$$\begin{aligned} \delta\epsilon_h &= C_V^{-1} \sum_m (\epsilon_m - \epsilon_{0w}) C_m \\ &= C_V^{-1} \sum_m \epsilon_m C_m - \epsilon_{0w} = \bar{\epsilon}_h - \epsilon_{0w}, \end{aligned} \quad (22)$$

$$\Delta(\mathbf{r}) = \frac{1}{\delta\epsilon_h C_V V_0} \sum_m (\epsilon_m - \epsilon_{0w}) U_m(\mathbf{r}),$$

$$\int \Delta(\mathbf{r}) d^3r = 1. \quad (23)$$

According to Ref. 27, moving-density fluctuations may be represented as quasiparticles named phonons.

Let us express the water density fluctuation through the properties of the phonon field. The continuity equation for density fluctuations is written as

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} + \text{div}[\dot{\mathbf{q}}(\mathbf{r}, t) \rho(\mathbf{r}, t)] = 0, \quad (24)$$

where $\mathbf{q}(\mathbf{r}, t)$ is a fluctuation coordinate. In the first approximation relative to $\delta\rho(\mathbf{r}, t)$, we have the following equation²⁷:

$$\frac{\delta\rho(\mathbf{r}, t)}{\rho_0} = -\nabla \mathbf{q}(\mathbf{r}, t) = -\frac{\phi(\mathbf{r}, t)}{u_0 \sqrt{\rho_0}}, \quad (25)$$

where u_0 is the velocity of the thermal fluctuation in water²⁹ (with the linear phonon dispersion law: $\omega_0(\mathbf{k}) = u_0 |\mathbf{k}|$, where \mathbf{k} is the phonon's momentum) and $\phi(\mathbf{r}, t)$ is a function that determines a high-frequency phonon field. By substituting Eqs. (19), (21), and (23) into Eq. (7), we have the following equation for the change of energy of water:

$$\begin{aligned} \delta E &= -\frac{\delta\epsilon_h C_V}{4\pi u_0 \sqrt{\rho_0}} \int d^3r \mathbf{E}(\mathbf{r}, t) \\ &\times \int d^3r' \Delta(\mathbf{r} - \mathbf{r}') \phi(\mathbf{r}', t) \mathbf{E}(\mathbf{r}', t), \end{aligned} \quad (26)$$

where

$$\Delta(\mathbf{r}) = \frac{\int_r^\infty f(r') dr'}{4\pi \int_0^\infty r'^2 dr' \int_{r'}^\infty f(r'') dr''}. \quad (27)$$

The hydrosol particles in this model are perceived as small potentials in a Brownian motion.³⁰⁻³⁴ They are characterized by the size distribution function $f(r)$ [$\int_0^\infty f(r) dr = 1$].

Let us express the electric field $\mathbf{E}(\mathbf{r}, t)$ through the potentials of the electromagnetic field:

$$\mathbf{E}(\mathbf{r}, t) = -\frac{1}{c} \frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t} - \text{grad}[\Phi(\mathbf{r}, t)], \quad (28)$$

where \mathbf{A} is the vector potential, Φ is the scalar potential, and c is the speed of light in vacuum. We accept the calibration with $\Phi = 0$. Such a model allows one to derive the operator of thermal density fluctuations $\hat{\psi}$. Let us introduce the following dimensionless parameters:

$$\begin{aligned} \psi(\mathbf{r}, t) &= \frac{1}{4\pi} \left(\frac{\bar{r}^3}{T} \right)^{1/2} \int d^3r' \Delta(\mathbf{r} - \mathbf{r}') \phi(\mathbf{r}', t) \\ &\equiv -\frac{\bar{r}}{4\pi} \left[\frac{\bar{r} u_0(T)}{\rho_0 T} \right]^{1/2} \int \Delta(\mathbf{r} - \mathbf{r}') \delta\rho(\mathbf{r}', t) d^3r', \end{aligned} \quad (29)$$

$$g = k_g C_V, \quad k_g = \delta\epsilon_h \left[\frac{T}{\bar{r}^3 \rho u_0^2(T)} \right]^{1/2} \approx 10^3 - 10^5, \quad (30)$$

where $\bar{r} = \int_0^\infty f(r) r dr$ is the mean radius of the hydrosol particles and T is the absolute temperature in kelvins.

Now we arrive at the formula for the interaction Hamiltonian between photons and thermal density fluctuations:

$$\hat{H}_{\text{int}}(t) = -\frac{g}{c^2} \int d^3r \frac{\partial \hat{A}_\alpha(\mathbf{r}, t)}{\partial t} \hat{\psi}(\mathbf{r}, t) \frac{\partial \hat{A}_\alpha(\mathbf{r}, t)}{\partial t}, \quad \alpha = 1, 2, 3, \quad (31)$$

where \hat{A}_α is a photon field operator that corresponds to a vector potential of the light wave and index α denotes a vector's component. Repeating indices everywhere in this paper imply summation.

4. GREEN'S FUNCTION

The Green's function of light in seawater with hydrosol particles is represented by the following equation [see Ref. 27, Eq. (28.7)]:

$$\begin{aligned} G_{\alpha\beta}(\tau_1 - \tau_2, \mathbf{r}_1, \mathbf{r}_2) &= -\text{Tr}\{\exp[(F - \hat{H}_{\text{int}})/T] \exp(\hat{H}_{\text{int}}|\tau_1 - \tau_2) \\ &\quad \times A_\alpha(\mathbf{r}_1) \exp(-\hat{H}_{\text{int}}|\tau_1 - \tau_2) A_\beta(\mathbf{r}_2)\}, \end{aligned} \quad (32)$$

where F is the free energy of the system and the symbol Tr (trace) denotes the summation over diagonal elements of the matrix. The photon Green's function $G_{\alpha\beta}$ in the space-energy representation satisfies the following wave equation derived from Maxwell's equations:

$$\begin{aligned} [\epsilon(\omega, \mathbf{r}) \omega^2 \delta_{\alpha\beta} - \text{curl}_{\alpha\gamma} \text{curl}_{\gamma\delta}] G_{\delta\beta}(\omega, \mathbf{r}, \mathbf{r}') &= 4\pi \delta_{\alpha\beta} \delta(\mathbf{r}_1 - \mathbf{r}_2). \end{aligned} \quad (33)$$

The Green's function $G_{\alpha\beta}^{(0)}(\tau_1 - \tau_2, \mathbf{r}_1, \mathbf{r}_2)$ of light in seawater without hydrosol particles is represented by Eq. (32) with $\hat{H}_{\text{int}} = 0$. The function $G_{\alpha\beta}^{(0)}$ in the space-energy representation satisfies the following wave equation:

$$\begin{aligned} [\epsilon_0(\omega, \mathbf{r}) \omega^2 \delta_{\alpha\beta} - \text{curl}_{\alpha\gamma} \text{curl}_{\gamma\delta}] G_{\delta\beta}^{(0)}(\omega, \mathbf{r}, \mathbf{r}') &= 4\pi \delta_{\alpha\beta} \delta(\mathbf{r}_1 - \mathbf{r}_2). \end{aligned} \quad (34)$$

The Green's function of photons propagating in a non-scattering medium with the dielectric permittivity ϵ_0 can be represented as a sum of the transverse and longitudinal components. In the energy-momentum representation it has the following form (see Ref. 28, Chap. 6):

$$G_{\alpha\beta}^{(0)}(\omega, \mathbf{k}) = G_0^{\text{tr}}(\omega, \mathbf{k})(\delta_{\alpha\beta} - n_\alpha n_\beta) + G_0^l(\omega, \mathbf{k}) n_\alpha n_\beta, \quad (35)$$

where

$$G_0^{\text{tr}}(\omega, \mathbf{k}) = \frac{4\pi}{\epsilon_0 \omega^2 / c^2 - k^2}, \quad G_0^l(\omega, \mathbf{k}) = \frac{4\pi c^2}{\epsilon_0 \omega^2}, \quad (36)$$

n_α is the component of the unity vector in the direction of \mathbf{k} , and $\delta_{\alpha\beta}$ is Kronecker's symbol or the unity tensor.^{26,28}

The Green's function of the photon field, which includes multiple scattering that is due to the interaction with thermal fluctuations described by the Hamiltonian (31), can be written as

$$G_{\alpha\beta}(\omega, \mathbf{k}) = G^{\text{tr}}(\omega, \mathbf{k})(\delta_{\alpha\beta} - n_\alpha n_\beta) + G^l(\omega, \mathbf{k}) n_\alpha n_\beta, \quad (37)$$

$$G^{\text{tr}}(\omega, \mathbf{k}) = \frac{4\pi}{\epsilon^{\text{tr}} \omega^2 / c^2 - k^2}, \quad G^l(\omega, \mathbf{k}) = \frac{4\pi c^2}{\epsilon^l \omega^2}, \quad (38)$$

where ϵ^{tr} and ϵ^l are, respectively, the transverse and longitudinal components of the dielectric permittivity tensor:

$$\epsilon_{\alpha\beta}(\omega, \mathbf{k}) = \epsilon^{\text{tr}}(\omega, \mathbf{k})(\delta_{\alpha\beta} - n_\alpha n_\beta) + \epsilon^l(\omega, \mathbf{k}) n_\alpha n_\beta. \quad (39)$$

It is clear from Eqs. (37)–(39) that the problem of finding the dielectric permittivity is equivalent to the problem of finding the Green's function.

Let us calculate the multiple-scattering Green's function and the corresponding dielectric permittivity. As a starting zero approximation, let us take the Green's function (35) that corresponds to the pure water. The dielectric permittivity of the clear water depends only on the circular frequency ω :

$$\epsilon_{\alpha\beta}^0(\omega) = \epsilon_0(\omega) \delta_{\alpha\beta} \equiv \epsilon_0(\omega)(\delta_{\alpha\beta} - n_\alpha n_\beta) + \epsilon_0(\omega) n_\alpha n_\beta. \quad (40)$$

It means that Eq. (40) takes into account only temporal dispersion that is determined by the processes of absorption and emission of photons by the water molecules.

The transverse and longitudinal components of the water dielectric permittivity that takes into account processes of multiple scattering on the hydrosol particles can be expressed as

$$\begin{aligned} \epsilon^{\text{tr}}(\omega, \mathbf{k}) &= \epsilon_0(\omega) + \delta\epsilon^{\text{tr}}(\omega, \mathbf{k}), \\ \epsilon^l(\omega, \mathbf{k}) &= \epsilon_0(\omega) + \delta\epsilon^l(\omega, \mathbf{k}), \\ \epsilon_0(\omega) &= \epsilon_0'(\omega) + i\epsilon_0''(\omega), \end{aligned} \quad (41)$$

where ϵ_0' and ϵ_0'' are the real and imaginary parts of the dielectric permittivity of pure water.

To calculate $\delta\epsilon^{\text{tr}}$ and $\delta\epsilon^l$, let us carry out the standard procedure²⁴ to calculate corrections to the Green's function starting from the Hamiltonian given by Eq. (31). The calculations are carried out on a discrete set of points distributed over the imaginary axis of the complex frequency $\hat{\omega} = \omega + i\omega_n$ ($\omega_n = 2\pi nT$, $n = 0, \pm 1, \pm 2, \dots$) in the system of units with $c = \hbar = 1$. This procedure cor-

responds to using the temperature Green's-function technique proposed by Matsubara.³⁵ This technique is valid when the temperature of seawater ($T_w \sim 300$ K) is much less than the temperature of light radiation ($T_{\text{rad}} \sim 30,000$ K). There is one important feature that simplifies this technique: the dispersion law of phonons is irrelevant. This feature is a consequence of the fact that the water mass in the fluctuating volume (phonon mass) is much larger than the photon mass. The scattering of the photon on a hydrosol particle pushed by the phonon occurs without a change of energy (elastic scattering). Only the *momentum* of the photon changes. This feature allows us to drop all summations over discrete frequencies. Further calculations can be carried out either analytically or with the help of Feynman diagrams.

The decomposition of the exponential terms in Eq. (32) into series and transition to the energy-momentum representation (Fourier transform) leads to the rules¹¹ in Subsection 4.A to calculate the Green's function of light in seawater with hydrosol particles by means of the Feynman diagram technique.

A. Rules for the Diagram Technique

The photon Green's function is represented as a dashed line, and the phonon Green's function is represented as a dotted line. A subscript (a Greek letter such as α or β) is associated with the end of each line. Each line has its own energy and momentum.

So let us form all connected topologically nonequivalent diagrams containing $2n$ circles and two external photon lines. All diagrams with an odd number of circles are equal to zero because the average of the product of an odd number of phonon operators, proportional to the density fluctuation, is equal to zero [see Eqs. (25) and (29)]. The circles of even diagrams are connected with the dotted (phonon) lines. An arbitrary diagram of $2n$ th order contains $3n - 1$ internal lines and $2n$ apexes denoted by circles. When the order of scattering is calculated, the following rules are satisfied:

1. Input and output photon lines correspond to the expression

$$\pm i\omega_n G_{\alpha\beta}^{(0)}(\omega_n, \mathbf{k}), \quad (42)$$

where

$$G_{\alpha\beta}^{(0)}(\omega_n, \mathbf{k}) = -\frac{4\pi(\delta_{\alpha\beta} - n_{\alpha}n_{\beta})}{\epsilon_0\omega_n^2 + \hbar^2} - \frac{4\pi n_{\alpha}n_{\beta}}{\epsilon_0\omega_n^2}. \quad (43)$$

2. An internal photon line corresponds to the expression

$$\omega_n^2 G_{\alpha\beta}^{(0)}(\omega_n, \mathbf{k}). \quad (44)$$

3. The internal phonon line is represented by

$$\frac{\bar{r}^3}{4\pi^2 T} \Delta^2(\mathbf{q}) D^{(0)}(0, \mathbf{q}) \cong -\frac{\bar{r}^3}{4\pi^2 T} \Delta^2(\mathbf{q}), \quad (45)$$

where

$$D^{(0)}(\omega, \mathbf{q}) = \frac{\omega_0^2(\mathbf{q})}{\omega^2 - \omega_0^2(\mathbf{q}) + i\delta},$$

$$\omega_0(\mathbf{q}) = u_0|\mathbf{q}|, \quad \Delta(\mathbf{q}) \equiv \int d^3r \exp(-i\mathbf{q}\mathbf{r}) \Delta(\mathbf{r}) \quad (46)$$

(δ is a very small positive value significant only when $\omega \neq 0$).

4. The result is multiplied by the factor

$$\left[-\frac{g^2 T}{(2\pi)^3} \right]^n. \quad (47)$$

5. The law of momentum conservation is satisfied in each vertex (circle). The energy or the frequency is transferred only over photon lines (elastic scattering).

B. Dyson's Equation

Using the rules presented in Subsection 4.A, we can write down the following Dyson's equation²⁷ for the Green's function of the photons in turbid water:

$$\begin{aligned} G_{\alpha\beta}(\omega_n, \mathbf{k}) &= G_{\alpha\beta}^{(0)}(\omega_n, \mathbf{k}) + G_{\alpha\gamma}^{(0)}(\omega_n, \mathbf{k}) \\ &\quad \times \pi_{\gamma\mu}(\omega_n, \mathbf{k}) G_{\mu\beta}(\omega_n, \mathbf{k}), \\ \pi_{\alpha\beta}(\omega_n, \mathbf{k}) &= -gh \int d^3\mathbf{q} \Delta^2(\mathbf{q}) G_{\alpha\gamma}(\omega_n, \mathbf{k} - \mathbf{q}) \\ &\quad \times \Gamma_{\gamma\beta}(\omega_n, \mathbf{k} - \mathbf{q}, \mathbf{k}), \\ h &= \bar{r}^3 \omega_n^4 / (2\pi)^5, \end{aligned} \quad (48)$$

where $\pi_{\alpha\beta}$ is the polarization operator and $\Gamma_{\alpha\beta}$ is the total vertex part that corresponds to the sum of all orders of photon scattering. The polarization operator is linked to the dielectric permittivity tensor by the equation

$$\delta\epsilon_{\alpha\beta}(\omega_n, \mathbf{k}) = \frac{4\pi}{\omega_n^2} \pi_{\alpha\beta}(\omega_n, \mathbf{k}). \quad (49)$$

Figure 1 shows the graphical form of Dyson's equation (48). The thick dashed lines represent the total photon Green's function. The triangle represents the total vertex part. In the derivation of Eq. (48), the effects of light scattering on itself (closed photon loops connected with the main photon line by phonon lines) are neglected. The input from these effects is extremely small. Only the diagrams with a single continuous photon line interacting with phonons are considered.

To calculate the vertex part, let us consider the perturbation series shown in Fig. 2. Because of the complexity of the diagrams' topology and the lack of a simple structure, it is impossible to formulate a simple integral equation for the vertex part. But close analysis of the perturbation series displayed in Fig. 2 shows that, among diagrams of the same order, the largest ones can be disassembled by cutting only two phonon lines. The photon Green's functions that correspond to these diagrams are paired together in such a manner that the poles of each Green's function merge when the transferred momentum $\mathbf{p} \rightarrow 0$. In Fig. 2 such diagrams are marked as a and c .

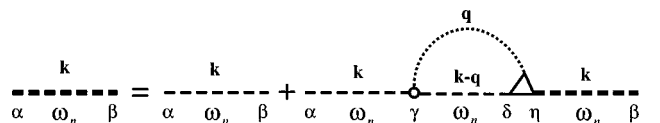


Fig. 1. Graphical form of Dyson's equation (48).

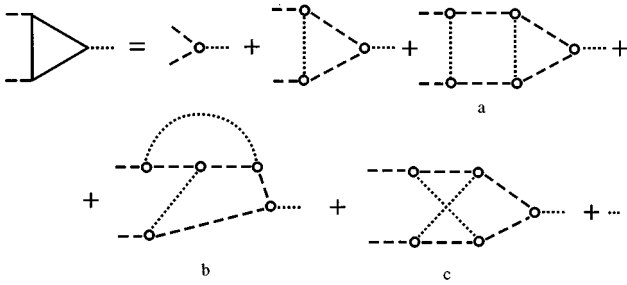


Fig. 2. Perturbation series for the vertex part $\Gamma_{\alpha\beta}$.

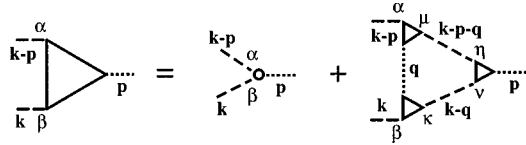


Fig. 3. Graphical form of the approximate integral equation (50) for the vertex part $\Gamma_{\alpha\beta}$.

By summing these largest diagrams, we have the equation for the total vertex part, shown in Fig. 3 as a diagram. It corresponds to the following integral equation for the vertex part:

$$\begin{aligned} \Gamma_{\alpha\beta}(\mathbf{k} - \mathbf{p}, \mathbf{k}) &= g \delta_{\alpha\beta} + h \int d^3\mathbf{q} \Delta^2(\mathbf{q}) \Gamma_{\alpha\mu}(\mathbf{k} - \mathbf{p}, \mathbf{k} - \mathbf{p} - \mathbf{q}) \\ &\quad \times G_{\mu\eta}(\mathbf{k} - \mathbf{p} - \mathbf{q}) \Gamma_{\eta\nu}(\mathbf{k} - \mathbf{p} - \mathbf{q}, \mathbf{k} - \mathbf{q}) \\ &\quad \times G_{\nu\kappa}(\mathbf{k} - \mathbf{q}) \Gamma_{\kappa\beta}(\mathbf{k} - \mathbf{q}, \mathbf{k}). \end{aligned} \quad (50)$$

To solve Eq. (50), we restrict ourselves to the scattering on large particles ($2\bar{r} \gg \lambda$), where λ is the wavelength of light. In this case $p < q < 1/\bar{r} \ll k$. By representing the vertex part as the sum of transverse and longitudinal parts, analogously to Eq. (39), integrating over momentum \mathbf{q} , and analytically expanding to the real frequency axis, we obtain the following system equations for the components of the vertex part:

$$\Gamma^{\text{tr}} = g + \frac{\bar{r}^2 \omega^4}{2\pi^2 k^2 c^4} (\Gamma^{\text{tr}})^3, \quad \Gamma^l = g + \frac{2}{3\pi^2 (\epsilon^l)^2} (\Gamma^l)^3. \quad (51)$$

5. DIELECTRIC PERMITTIVITY

Let us derive equations that link corrections to the dielectric permittivity with the vertex part. Using Eqs. (31), (40), (48), and (49), integrating over momentum \mathbf{q} , and analytically transferring to the real frequency axis, we have the following equations:

$$\epsilon^{\text{tr}} = \epsilon_0 + ig \frac{\bar{r}\omega|\omega|}{16\pi kc^2} \Gamma^{\text{tr}}, \quad \epsilon^l = \epsilon_0 - \frac{g}{6\pi^2 \epsilon^l} \Gamma^l. \quad (52)$$

To have a closed system of equations, we add the dispersion relation taken from Maxwell's equations:

$$k^2 = \epsilon^{\text{tr}} \omega^2 / c^2. \quad (53)$$

Now we have two complete systems of equations for the complex parameters ϵ^{tr} and ϵ^l :

$$\epsilon^l = \epsilon_0 - \frac{g}{6\pi^2 \epsilon^l} \Gamma^l, \quad \Gamma^l = g + \frac{2}{3\pi^2 (\epsilon^l)^2} (\Gamma^l)^3, \quad (54)$$

$$k^2 = \epsilon^{\text{tr}} \frac{\omega^2}{c^2}, \quad \epsilon^{\text{tr}} = \epsilon_0 + \frac{igx}{16\pi \sqrt{\epsilon^{\text{tr}}}} \Gamma^{\text{tr}},$$

$$\Gamma^{\text{tr}} = g + \frac{x^2}{2\pi^2 \epsilon^{\text{tr}}} (\Gamma^{\text{tr}})^3, \quad (55)$$

where $x = \bar{r}|\omega|/c = 2\pi\bar{r}/\lambda$ is the size parameter.³⁶ In the general case of arbitrary values of x and g (which corresponds to arbitrary concentrations), the systems of equations (54) and (55) have no analytic solutions. For the value of the parameter

$$g < 4\sqrt{2}\epsilon'_0/3, \quad C_V < 2 \times 10^{-3} \quad (\text{or } C_p < 2 \text{ g/m}^3), \quad (56)$$

we have an approximate solution for the longitudinal component of the dielectric permittivity tensor:

$$\begin{aligned} \epsilon^l &= \epsilon'_0 \left\{ 1 - \frac{\eta}{3} \sin \left[\frac{1}{3} \sin^{-1}(3\eta) \right] \right\}, \\ \eta &= \frac{C_V \delta \epsilon_h}{\pi \epsilon'_0} \left(\frac{T}{2\bar{r}^3 \rho u_0^2} \right)^{1/2}, \end{aligned} \quad (57)$$

where $C_p \equiv \rho C_V$ is the concentration of hydrosol particles in conventional units (g/m^3). According to Eqs. (54), ϵ^l is determined only by the water properties. As is seen from Eqs. (54), this is also true for any water turbidity.

6. SCATTERING COEFFICIENT

Let us write an expression that connects the transverse part of the dielectric permittivity ϵ^{tr} with the beam scattering coefficient b . According to wave theory and Bouguer's law, the absolute value of the photon wave vector k is

$$k = k_0 + i(a + b)/2; \quad (58)$$

at the same time, from the dispersion relationship we have

$$k = \frac{\omega \sqrt{\epsilon^{\text{tr}}}}{c}. \quad (59)$$

By introducing an auxiliary variable

$$z = i\sqrt{\epsilon^{\text{tr}}} \quad (60)$$

and taking into account that the absorption coefficient a is due to absorption processes in background water, which are not included in our calculations, we have

$$b = \frac{4\pi}{\lambda} \text{Re}(z) \equiv \frac{4\pi}{\lambda} \text{Im}(\sqrt{\epsilon^{\text{tr}}}). \quad (61)$$

By solving Eqs. (55) with respect to ϵ^{tr} and replacing it with the variable z according to Eq. (60), we have the following algebraic equation of seventh order with respect to z :

Table 1. Values of the Parameter $Re(z)$ for Different Size Parameters $x = (2\pi\bar{r})/\lambda$ and Concentrations

g	x			
	20	200	2000	20,000
0.001	0.0000382520083981352	0.00003889708867014887	0.00004405591503813641	0.00006808675127872522
0.01	0.0003889968127614913	0.0004413463309078073	0.0006808672289176459	0.001331001567394975
0.1	0.004413414061668614	0.006808408679003452	0.01330778093474727	0.02801921767710799
0.3	0.01560196002156826	0.02763816807094165	0.05656906619476877	0.119261899074067
0.5	0.028843048657225	0.05355156834986856	0.1103724238115074	0.2273210624193799
1.0	0.06782403253850639	0.130962807451096	0.2629058831299653	0.4915729190820884
3.0	0.2607548913473402	0.4718230761749745	0.7903553580963729	1.221217048860729
5.0	0.4547320997689933	0.7537526964658059	1.16805018412666	1.722581232924349
10	0.8461096101753952	1.275010732778465	1.85970893251966	2.653578357080784
20	1.401622773034389	2.010392007934948	2.851179681305613	4.007903143447798
40	2.18674273530717	3.067821116464854	4.296750683666268	5.999602204397481
80	3.318417336257999	4.612472826798015	6.425117946818375	8.944848607997034
160	4.973479661691648	6.888735539322619	9.574155923744264	13.31161745998768
320	7.413249143693385	10.25776056977357	14.24414545197939	19.79389237565583
640	11.02398143083176	15.25446521655215	21.17714270651046	29.42191539762678
1280	16.37789542378423	22.67241926639643	31.47487451805124	43.72591945758872
2560	24.3239651017216	33.68991934073195	46.77382967672264	64.97934212108541
5120	36.12275211972931	50.05723270267769	69.50535480801077	96.56017019972301
10,000	52.9255369717243	73.37394015133673	101.8919179243287	141.5563893122459

Table 2. Regression Coefficients for Equation (63)

x	β_1	β_2	β_3	α	γ	g_0
20	0.09806933	-0.001680836	0.00001488722	0.266087787	0.5754037	40
200	0.16837140	-0.004626691	0.00006162638	0.365200048	0.576702	20
2000	0.27688820	-0.007127919	-0.00019729570	0.506416969	0.577014	10
20,000	0.49303440	-0.025888510	-0.00076870260	0.704132733	0.5770671	5

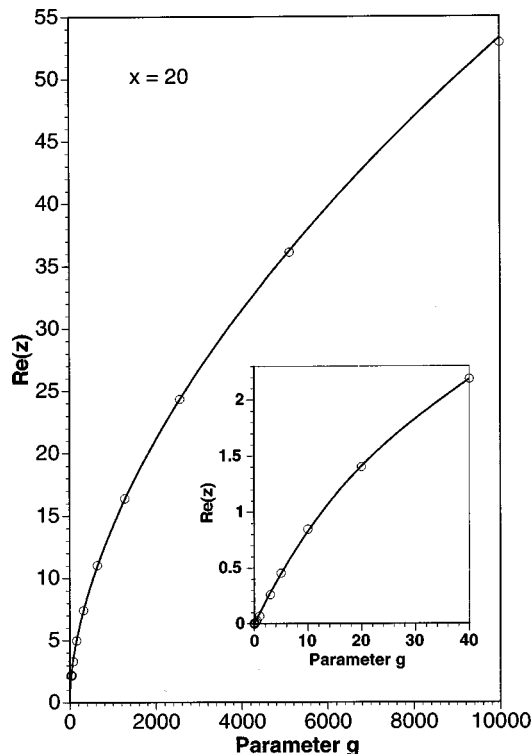


Fig. 4. Concentration dependence of the relative scattering coefficient $Re(z)$ for size parameter $x = (2\pi\bar{r})/\lambda = 20$.

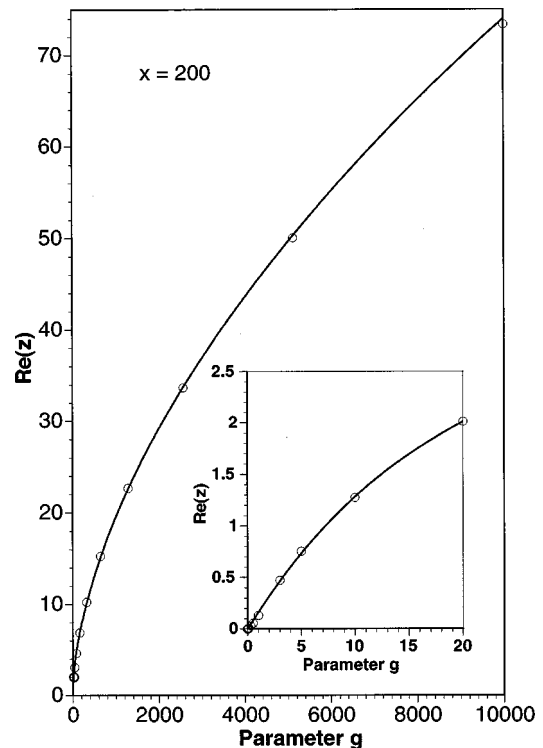


Fig. 5. Same as Fig. 4, but for $x = 200$.

$$16\pi z(\epsilon_0 + z^2)[g^2 + 128(\epsilon_0 + z^2)^2] - g^4 x = 0. \quad (62)$$

Equation (62) has no analytic solutions. A numerical analysis shows that it has one real and six complex roots. There is only one complex physical root of Eq. (62).³⁷ The real part of this physical root, which can be used for cal-

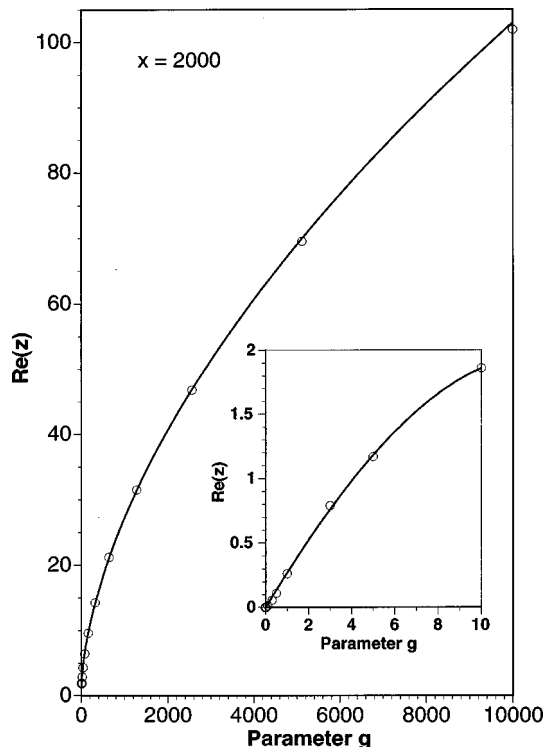


Fig. 6. Same as Fig. 4, but for $x = 2000$.

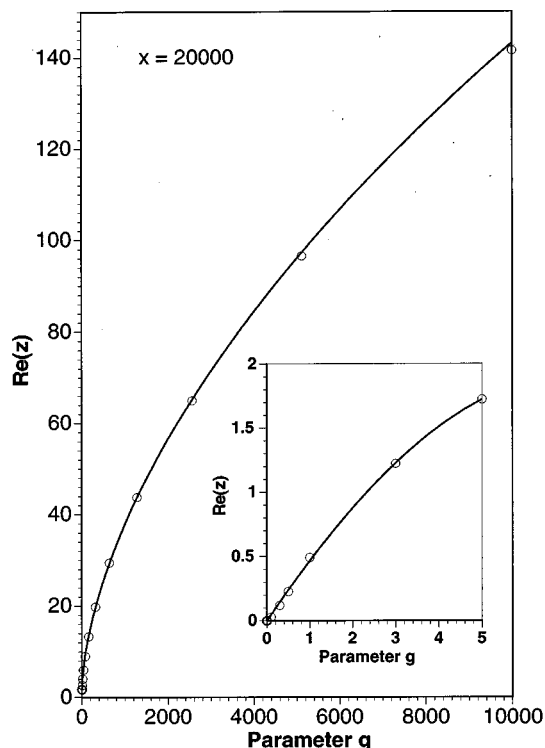


Fig. 7. Same as Fig. 4, but for $x = 20,000$.

ulation of the scattering coefficient with Eq. (61), is computed for different sets of parameters x and g and included in Table 1.

Using values given in Table 1, we obtain the following regressions, which express the scattering coefficient as a function of the concentration parameter $g \sim C_p$:

$$b = \frac{4\pi}{\lambda} \begin{cases} \beta_1 g + \beta_2 g^2 + \beta_3 g^3, & g < g_0 \\ \alpha g^\gamma, & g \geq g_0 \end{cases}, \quad r^2 > 0.999. \quad (63)$$

The values of parameters β_i ($i = 1, 2, 3$), α , γ , and g_0 for different values of the size parameter are given in Table 2.

Figures 4–7 show the concentration dependence of parameter $\text{Re}(z) = b\lambda/(4\pi)$ for different values of the size parameter x .

At very small concentrations of particles, the dependence $b(g) \equiv b(C_p) = (4\pi/\lambda)\text{Re}[z(C_p, x)]$ is linear in concentration (g or C_p). At higher concentrations this dependence is similar to the experimental dependence proposed by Prieur and Sathyendranath.⁴

7. CONCLUSION

It is shown that the approach based on Maxwell's theory in a stochastically scattering medium can be productively used in ocean optics. The dielectric permittivity tensor of water with imbedded scattering particles is found as a solution of Dyson's equation. Starting from the equations for the dielectric permittivity, a nonlinear dependence of the scattering coefficient of water is derived. It is shown that the scattering coefficient of water depends nonlinearly on the concentration of scatterers. The dependence is linear when the concentration of scattering particles is very small. At higher concentrations the concentration dependence weakens and can be represented by a power law: $b \sim C_p^{0.576}$.

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