Critical-opalescence saturation in multiple scattering of light

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Multiple scattering of an electromagnetic wave by a volume of liquid near the critical point is considered within the framework of a macroscopic approach in which the dielectric-constant fluctuations are assumed to be due to density fluctuations. The mean electric field strength and the coherence tensor are determined by using the exact Dyson and Bethe-Salpeter equations, whose kernels (the mass operator and the intensity operator) are related, via the mean Green's tensor, by the optical theorem in the theory of multiple scattering of waves. An exact equation is derived for the incoherent-scattering cross section, integrated over all angles, in terms of effective dielectric tensor of a randomly inhomogeneous medium and of the mean electric field. The calculation and the aid of this equation is carried out in an approximation wherein the effective complex refractive index differs little from unity. It is shown that the integral cross section for incoherent scattering near the critical point is bounded from above by the maximum geometric transverse section of the volume of the material.

1. INTRODUCTION

Critical opalescence is defined[1] as the abrupt increase of scattered-light intensity, predominantly in the propagation direction of the exciting light, near the critical point of an individual substance or near the critical stratification or mixing temperature of binary solutions. The extinction coefficient, calculated according to the Ornstein-Zernike theory for a liquid in the single-scattering approximation, diverges logarithmically at the critical point itself, owing to the contribution of the small scattering angles. No such difficulty arises in real systems, since the correlation length of the density fluctuations is restricted by the vessel dimensions.[2] The divergence is eliminated also when account is taken of diffraction by the volume of the liquid.[3] The extinction coefficient at the crucial point, however, is restricted also by another factor, namely the multiple scattering of the light.[4]

The question of the value of the extinction coefficient h in multiple scattering of light in a liquid that is in a near-critical state was considered by Lakoza and Chalyĭ.[5,6] They obtained by a macroscopic approach an equation [1)] in the form

$$h=h_1/(1-h_1L_0), \quad h_1L_0<1, \tag{1}$$

where h_1 is the extinction coefficient in the single-scattering approximation, and L_0 is the characteristic dimension of the scattering volume. Equation (1) does not include the aforementioned saturation of the extinction coefficient at the critical point, in view of the multiple scattering. Moreover, the extinction coefficient should have according to (1) an additional divergence as the critical point is approached, when $h_1L_0\to 1$. The cause of these shortcomings of Eq. (1) is easily explained. The equation was derived in fact by using the Bethe-Salpeter equation in the ladder approximation.[7] In Refs. 5 and 6, however, no account was taken of the contribution of the extinction effect to the value of the bilinear combination of the Green's function averaged over the ensemble, so that the energy-flux conservation law in multiple wave scattering was violated.

We derive here an exact equation for the multiple-scattering extinction coefficient h, starting from the exact Dyson and Bethe-Salpeter equations.[8] The decisive factor in the derivation is the use of the optical theorem in the theory of multiple scattering of waves,[9,10] expressing the imaginary part of the mass operator in terms of the imaginary part of the mean Green's function and the intensity operator. The exact equation represents the extinction coefficient h as a double integral, over the scattering volume, of the product two quantities. One is the imaginary part of the effective-dielectric constant of a randomly inhomogeneous medium,[11] and the other a bilinear combination of the mean electric field. According to this representation the extinction coefficient coincides, in accord with its definition and the energy-flux conservation law, with the cross section for the absorption of the coherent radiation (of the mean field)[12] per unit volume. In a randomly inhomogeneous medium without true absorption, coherent-radiation absorption is taken to mean energy-flux transfer from the mean field to the fluctuating component of the field—to the incoherent radiation. The extinction coefficient is calculated by means of the exact formula in an approximation using an effective complex refractive index for the mean field,[2] and neglecting, according to Hulst,[13] reflection and refraction of the mean field at the boundary of the scattering volume. It is found therefore that the coefficent of extinction of multiply scattered light tends at the critical point to the ratio of the geometric cross section of the scattering volume to the size of the volume. This means that the scattering volume behaves at the critical point as a "black body" for the coherent radiation, converting all of the incident coherent radiation into incoherent.

2. INITIAL EQUATIONS

In the macroscopic approach[6] to light scattering in a substance (liquid) in near-critical state, use is made of the concept of the random dielectric constant $\varepsilon(\mathbf{r})=\varepsilon_0+\tilde{\varepsilon}(\mathbf{r})$. Here $\varepsilon_0=\langle\varepsilon(\mathbf{r})\rangle$ is the dielectric constant of the medium averaged over an ensemble of realizations (we assume this

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mean value to be constant and equal to the dielectric constant of the homogeneous medium), and $\tilde{\varepsilon}(\mathbf{r})$ is its fluctuating part. Assume that a plane monochromatic electromagnetic wave with an electric field

$$\mathbf{E}_0(\mathbf{r})=\mathbf{E}_0\exp(ik_0\mathbf{n}_0\mathbf{r}),\quad \mathbf{E}_0\mathbf{n}_0=0,\qquad k_0=(\omega/c)\,\varepsilon_0^{1/2} \tag{2}$$

is incident on the volume in the direction of a unit vector $\mathbf{n}_0$. The mean electric field $\langle E_\mu(\mathbf{r})\rangle$ and its mutual coherence tensor $\langle E_\mu(\mathbf{r})E^*_{\mu'}(\mathbf{r}')\rangle$, (the asterisk denotes the complex conjugate) can be obtained the Dyson and Bethe-Salpeter equations,[8] whose symbolic operator form is

$$\langle G\rangle=G_0+G_0M\langle G\rangle, \tag{3}$$

$$\langle E\times E^*\rangle=\langle E\rangle\times\langle E^*\rangle+\langle G\rangle\times\langle G^*\rangle K\langle E\times E^*\rangle, \tag{4}$$

$$E=\frac{4\pi\omega}{ic^2}Gj. \tag{5}$$

In this notation, the symbol $\times$ denotes the tensor product[14] of the vector functions or of the tensor kernels of the operators $G_{\mu\mu'}(\mathbf{r},\mathbf{r}')$ and $G^0_{\mu\mu'}(\mathbf{r},\mathbf{r}')$ which are the Green's tensors of the electric field with and without allowance for the fluctuations of the dielectric constant of the substance. The mass operator M and the intensity operator K are specified by their kernels $M_{\mu\mu'}(\mathbf{r},\mathbf{r}')$ and $K_{\mu\mu',\nu\nu'}(\mathbf{r}_1,\mathbf{r}_1';\mathbf{r}_2,\mathbf{r}_2^*)$, with the intensity operator acting on a certain test tensor $\Phi_{\mu\nu}(\mathbf{r}_1,\mathbf{r}_2)$ in accordance with

$$(K\Phi)_{\mu\nu}(\mathbf{r}_1,\mathbf{r}_2)=\iint d\mathbf{r}_1'\,d\mathbf{r}_2'K_{\mu\mu',\nu\nu'}(\mathbf{r}_1,\mathbf{r}_1';\mathbf{r}_2,\mathbf{r}_2')\,\Phi_{\mu'\nu'}(\mathbf{r}_1',\mathbf{r}_2'),$$

with summation over repeated indices; $j_\mu(\mathbf{r})$ is the current-density vector [the source of the incident field (2)].

The operators M and K are specified in the Feynman diagram technique by infinite sums of highly connective diagrams,[7] summation over which is impossible. General physical requirements, however, call for the kernels M and K to satisfy certain relations. These include the optical theorem in the theory of multiple scattering of waves[9,10]

$$M\otimes 1-1\otimes M^*=(\langle G\rangle\otimes 1-1\otimes\langle G^*\rangle)K. \tag{6}$$

Here $A\otimes B$ denotes the contracted tensor product of the nuclei $A_{\mu\mu'}(\mathbf{r},\mathbf{r}')$ and $B_{\mu\mu'}(\mathbf{r},\mathbf{r}'')$, equal to

$$\int d\mathbf{r}A_{\mu\mu'}(\mathbf{r},\mathbf{r}')B_{\mu\nu'}(\mathbf{r},\mathbf{r}''); \tag{7}$$

1 in (6) stands for the unit operator $\delta_{\mu\mu'}\delta(\mathbf{r}-\mathbf{r}')$.

Scattering of an electromagnetic wave by a volume of matter is described by the electric-field scattering operator $T_{\mu\mu'}(\mathbf{r},\mathbf{r}')$ defined by the relation,

$$E=E_0+G_0TE_0 \tag{8}$$

and satisfying an analog of the Lippmann-Schwinger equation[15] with an effective scattering potential $V(\mathbf{r})=-k_0^2\tilde{\varepsilon}(\mathbf{r})/\varepsilon_0$. Incoherent wave scattering is characterized by fluctuation of the scattering operator

$$U=\langle T\times T^*\rangle-\langle T\rangle\times\langle T^*\rangle. \tag{9}$$

We introduce the Fourier transform of the kernel of operator (9), putting

$$U_{\mu\mu',\nu\nu'}(\mathbf{p}_1,\mathbf{p}_1';\mathbf{p}_2,\mathbf{p}_2')=\int\ldots\int d\mathbf{r}_1\,d\mathbf{r}_1'\,d\mathbf{r}_2\,d\mathbf{r}_2'\times\exp[-i(\mathbf{p}_1\mathbf{r}_1-\mathbf{p}_1'\mathbf{r}_1')]\times\exp[i(\mathbf{p}_2\mathbf{r}_2-\mathbf{p}_2'\mathbf{r}_2')]U_{\mu\mu',\nu\nu'}(\mathbf{r}_1,\mathbf{r}_1';\mathbf{r}_2,\mathbf{r}_2'). \tag{10}$$

The differential cross section for incoherent scattering of the wave in the direction of the unit vector $\mathbf{n}$ by the volume of matter takes then the form

$$C_{\text{inc}}(\mathbf{n},\mathbf{n}_0)\equiv\frac{1}{(\mathbf{\Pi}_0\mathbf{n}_0)}\lim_{r\to\infty}r^2(\mathbf{\Pi}_{\text{inc}}\mathbf{n})=$$

$$=\frac{1}{(4\pi)^2}\frac{1}{(\mathbf{E}_0\mathbf{E}_0^*)}P_{\mu\nu}^{tr}(\mathbf{n})U_{\mu\mu',\nu\nu'}(k_0\mathbf{n},k_0\mathbf{n}_0;k_0\mathbf{n},k_0\mathbf{n}_0)E_{\mu'}{}^0E_{\nu'}{}^{0*}. \tag{11}$$

In the first equality $\mathbf{\Pi}_{\text{inc}}$ denotes the Poynting vector of the incoherent radiation

$$\mathbf{\Pi}_{\text{inc}}=\frac{c}{8\pi}\operatorname{Re}\langle[\tilde{\mathbf{E}}\tilde{\mathbf{H}}^*]\rangle,\quad \tilde{\mathbf{E}}=\mathbf{E}-\langle\mathbf{E}\rangle,\quad \tilde{\mathbf{H}}=\mathbf{H}-\langle\mathbf{H}\rangle, \tag{12}$$

and $\mathbf{\Pi}_0$ is the Poynting vector of the incident wave (2); the origin $r=0$ is inside the volume in the second equality, where

$$P_{\mu\nu}{}^{tr}=\delta_{\mu\nu}-n_\mu n_\nu.$$

Integrating (11) over all the scattering directions $\mathbf{n}$ we obtain the integral cross section for the incoherent scattering C_{inc}. With the aid of the equation

$$C_{\text{coh.abs.}}=-\frac{1}{(\mathbf{\Pi}_0\mathbf{n}_0)}\lim_{r\to\infty}r^2\int d^2\mathbf{n}(\mathbf{\Pi}_{\text{coh}}\mathbf{n}),\qquad \mathbf{\Pi}_{\text{coh}}=\frac{c}{8\pi}\operatorname{Re}[\langle\mathbf{E}\rangle\langle\mathbf{H}\rangle] \tag{13}$$

we introduce the cross section for the absorption of the coherent radiation $C_{\text{coh.abs.}}$. Subject to the condition that the random dielectric constant is real, the integral cross section for incoherent scattering coincides with the cross section for absorption of coherent radiation[12]

$$C_{\text{inc}}=C_{\text{coh.abs.}} \tag{14}$$

The basic relations of the present section, meaning the second equation of (11) and Eq. (14), can be easily verified. The coefficient of extinction of multiple scattering of light by a volume of matter is defined by the integral coherent-scattering section per unit volume Ω:

$$h=C_{\text{inc}}/\Omega. \tag{15}$$

Neglecting single scattering, this definition coincides with the customary one.[1]

3. EXPRESSION FOR EXTINCTION COEFFICIENT IN TERMS OF THE EFFECTIVE DIELECTRIC TENSOR

It follows from (8) and (9) that the fluctuation of the electric field is expressed in terms of the fluctuation of the scattering operator. Taken together with Eq. (4), this yields

$$(G_0\times G_0^*)U(E_0\times E_0^*)=\langle G\rangle\times\langle G^*\rangle K\langle E\times E^*\rangle. \tag{16}$$

We act on (16) from the left with the operator $G_0^{-1}\times 1-1\times G_0^{*-1}$ and carry out the contraction (7) of the tensor products. Taking (3) into account, we get

$$(1\otimes G_0^*-G_0\otimes 1)\,U(E_0\times E_0^*)=[1\otimes\langle G^*\rangle-\langle G\rangle\otimes 1$$
$$+(M\otimes 1-1\otimes M^*)\langle G\rangle\times\langle G^*\rangle]\,K\langle E\times E^*\rangle. \quad (17)$$

The first two terms on the right are transformed, after expanding the expression in the square brackets, with the aid of the optical theorem (6). Using next again Eq. (4), we ultimately get

$$(1\otimes G_0^*-G_0\otimes 1)\,U(E_0\times E_0^*)=(1\otimes M^*-M\otimes 1)\langle E\rangle\times\langle E^*\rangle. \quad (18)$$

The left-hand side of this relation is expressed with the aid of the Green's tensor of the electric field in a homogeneous medium[11]

$$G_{\mu\nu}^0(\mathbf{r})=-\left(\delta_{\mu\nu}+\frac{1}{k_0^2}\frac{\partial^2}{\partial x_\mu\,\partial x_\nu}\right)\frac{\exp(ik_0r)}{4\pi r} \quad (19)$$

and with the aid of the Fourier transform (10) in terms of the integral incoherent-scattering cross section C_{inc}. The right-hand side of (18) contains the imaginary part of the mass operator

$$\operatorname{Im}M=\frac{1}{2i}(M-M^+), \quad (20)$$

where the supscript + denotes the Hermitian adjoint operator. In view of the foregoing remarks, relation (18) is rewritten in the form

$$C_{\text{inc}}=-\frac{1}{k_0(\mathbf{E}_0\mathbf{E}_0^*)}\iint d\mathbf{r}_1\,d\mathbf{r}_2\,(\operatorname{Im}M)_{\mu\nu}(\mathbf{r}_1,\mathbf{r}_2)$$
$$\times\langle E_\nu(\mathbf{r}_2)\rangle\langle E_\mu^*(\mathbf{r}_1)\rangle. \quad (21)$$

The double integral in the right-hand side of (21) is a quadratic form of the self-adjoint operator (20). It follows therefore that this operator must be negative-definite:

$$\operatorname{Im}M<0. \quad (22)$$

The effective dielectric tensor $\varepsilon_{\mu\nu}^{\text{eff}}(\mathbf{r},\mathbf{r}')$ of a randomly inhomogeneous medium is introduced by the relation $\langle\varepsilon E\rangle=\varepsilon_{\partial\Phi\Phi}\langle E\rangle$ (Ref. 11). Writing for the field-strength vector of the electric field $E_\mu(\mathbf{r})$ the analog of the Lippmann-Schwinger equation[15] and averaging it over the ensemble of the fluctuations of the dielectric constant of the medium, we obtain

$$\varepsilon_{\mu\nu}^{\text{eff}}(\mathbf{r},\mathbf{r}')/\varepsilon_0=\delta_{\mu\nu}\delta(\mathbf{r}-\mathbf{r}')-k_0^{-2}M_{\mu\nu}(\mathbf{r},\mathbf{r}'). \quad (23)$$

This and (21) lead to an expression for the integral incoherent-scattering cross section in terms of the effective dielectric tensor of the medium and the mean electric field.

4. EFFECTIVE-REFRACTIVE-INDEX APPROXIMATION

We turn now to an approximate calculation of the double integral (21). We assume that the fluctuations of the dielectric constant of the medium are homogeneous and isotropic. In the interior of the medium the mass operator depends then only on the difference of its arguments, and its Fourier transform can be resolved into a sum of transverse and longitudinal components[11]

$$M_{\mu\nu}(\mathbf{p})=\int\exp(-i\mathbf{pr})\,M_{\mu\nu}(\mathbf{r})\,d\mathbf{r}$$
$$=P_{\mu\nu}^{tr}(\mathbf{p})\,M_t(p)+P_{\mu\nu}^{l}(\mathbf{p})\,M_l(p),\quad P_{\mu\nu}^{l}(\mathbf{p})=p_\mu p_\nu/p^2. \quad (24)$$

The effective complex refractive indices m_t and m_l of the tranverse and longitudinal components of the mean electric field inside the medium are determined with the aid of $M_t(p)$ and $M_l(p)$ by the dispersion relations

$$m_t^2=1-M_t(m_tk_0)/k_0^2,\quad M_l(m_lk_0)/k_0^2=1. \quad (25)$$

The same dispersion relations yield the poles of the Fourier transform of the mean Green's tensor of the electric field in an unbounded randomly inhomogeneous medium.[11] In the solution of the first dispersion equation in (25) we assume that the refractive index differs little from unity. Then

$$m_t\approx 1-{}^1/_2k_0^{-2}M_t(k_0),\quad |M_t(k_0)|/k_0^2\ll 1. \quad (26)$$

The quantity

$$h_1=-k_0^{-1}\operatorname{Im}M_t(k_0), \quad (27)$$

as will be made clear below, coincides with the extinction coefficient of the volume of matter in the single-scattering approximation. Under condition (26) the mean electric field inside the scattering volume can be calculated in the geometric-optics approximation, assuming according to Hulst[13] (see p. 202 of the Russian translation) that

$$\langle E_\mu(\mathbf{r})\rangle\approx E_\mu^0\exp(ik_0m_t\mathbf{n}_0\mathbf{r}). \quad (28)$$

We substitute[2)] (28) in the integer of (21). This leads, after simple calculations, with allowance for (15) and (27), to the following expression for the extinction coefficient of multiple scattering of electromagnetic wave (2) by a convex volume of matter

$$h=\frac{1}{\Omega}\int d\mathbf{r}_\perp[1-\exp(-h_1L(\mathbf{r}_\perp))]. \quad (29)$$

The integration here is over the maximum volume transverse section perpendicular to the wave-incidence direction $\mathbf{n}_0$; $L(\mathbf{r}_\perp)$ is the length from the entrance to and exit of the volume through which the beam passes in the $\mathbf{n}_0$ direction through the point $\mathbf{r}_\perp$ of the maximum transverse section.

5. SATURATION OF EXTINCTION COEFFICIENT AT THE CRITICAL POINT

It can be seen from (29) that the extinction coefficient h does not exceed the value

$$h\leqslant C_{\text{geom}}/\Omega, \quad (30)$$

where C_{geom} is the maximum geometric transverse section of the volume. This limiting value is reached under the condition[3)] $h_1L\gg 1$, when the scattering volume behaves as a black body with respect to the coherent radiation. In this opposite limiting case of single scattering, $h_1L\ll 1$, the extinction coefficient $h\approx h_1$. The figure shows a plot of the ratio h/h_1 vs the "optical thickness" parameter h_1L of a spherical volume of radius L, as calculated from (29). The figure shows also the analogous plot (upper curve) in accordance with Eq. (1).[4)] It can be seen that the plots differ in shape.

To calculate the single-scattering extinction coefficient h_1 we choose a mass operator in the lowest order of perturbation theory (the Bourret approximation, see Ref. 7, p. 403)

$$M_{\mu\nu}(\mathbf{r})=\langle V(\mathbf{r})\,V(\mathbf{r}'=0)\rangle G_{\mu\nu}^0(\mathbf{r}). \quad (31)$$

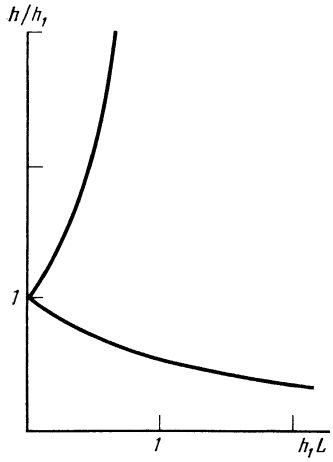

FIG. 1. Dependence of the ratio h/h_1 on the "optical thickness" parameter h_1L of a spherical volume of radius L.

According to Refs. 5 and 6, the effective scattering potential of the medium is

$$V(\mathbf{r}) \approx -\left(\frac{\omega}{c}\right)^2 \left(\rho_0 \frac{\partial \varepsilon_0}{\partial \rho_0}\right)_T \Delta\rho(\mathbf{r}), \qquad \Delta\rho(\mathbf{r}) = (\rho(\mathbf{r}) - \rho_0)/\rho_0, \tag{32}$$

where the partial derivative is taken at constant temperature T, $\rho(\mathbf{r})$ is the macroscopic density of the material, and ρ_0 is its mean value. On the basis of (31) and (32) we get from (27)

$$h_1 = \frac{1}{2(4\pi)^2}\left(\frac{\omega}{c}\right)^4 \left(\rho_0 \frac{\partial \varepsilon_0}{\partial \rho_0}\right)_T^2 \times \int d^2n\,[1+(\mathbf{n}\mathbf{n}_0)^2]\, g_2(k_0(\mathbf{n}-\mathbf{n}_0)). \tag{33}$$

We have denoted by $g_2(\mathbf{k})$ the density-fluctuation correlation function[5,6] that is connected with the static structure factor $S(\mathbf{k})$ by the simple relation (see Ref. 16, p. 285)

$$g_2(\mathbf{k}) = \int d\mathbf{r} \exp(-i\mathbf{k}\mathbf{r}) \langle \Delta\rho(\mathbf{r})\Delta\rho(0)\rangle = \frac{1}{\rho_0} S(\mathbf{k}). \tag{34}$$

We choose the structure factor in accord with the Ornstein-Zernike theory of critical fluctuations,[16] $S(k) = \xi_0^{-2}/(\xi^{-2} + k^2)$, where ξ is the correlation length and ξ_0 is the direct-interaction radius. According to (33), h_1 assumes then a known value[5,6] that yields for the optical thickness of the substance

$$h_1 L = \pi s \left[\frac{1+2\delta+2\delta^2}{\delta^2}\ln(1+2\delta) - \frac{2(1+\delta)}{\delta}\right], \qquad s = \frac{1}{2(4\pi)^2}\left(\frac{\omega}{c}\right)^4 \left(\rho_0 \frac{\partial \varepsilon_0}{\partial \rho_0}\right)_T^2 \frac{L}{(k_0\xi_0)^2 \rho_0}. \tag{35}$$

The parameter $\delta = 2(k_0\xi)^2$ increases without limit at the critical point, and the parameter s remains finite.

Let us determine the values of $k_0\xi$ starting with which multiple scattering of light near the critical point becomes substantial at a level $h/h_1 \leqslant 0.5$. From the lower plot of the figure we find $h_1L \geqslant 1.1$, which yields at $s = 0.1$, on the basis of (35), the value $k_0\xi \geqslant 1.7$ [see the table of h_1L/s as a function of δ in Ref. 5].

6. CONCLUSION

It is easily seen that the basic formula (21), with account taken of (23), can be obtained also without using (4), by merely determining the tensor $\varepsilon_{\mu\nu}^{\text{eff}}(\mathbf{r},\mathbf{r}')$ and the electromagnetic-energy flux conservation law. The transition from (21) to (29) is made under the assumptions customarily used on going from the Bethe-Salpeter equation to the radiation-transport equation.[12] This is due, in particular, to the condition $\xi h_1 \ll 1$. Equations (21) and (29) can be generalized to the case when the mean values of the dielectric constant $\varepsilon_0(\mathbf{r})$ and of its fluctuation $\bar{\varepsilon}(\mathbf{r})$ are spatially homogeneous and behave as tensors. This permits allowance for the effect of the gravitational field for the vapor + liquid system,[6] and consideration, besides critical opalescence of liquids and binary solutions, of similar phenomena such as enhancement of the scattered-light intensity near the temperature of a second-order phase transition,[17] and in liquid crystals.[18]

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APPENDIX

We indicate the assumptions under which Eq. (1) is obtained in the approach described in this paper.

We verify first satisfaction of the electromagnetic-field energy-flux conservation law for the solution of Eq. (4). We note for this purpose that the Green's function (19) of an electric field in a homogeneous medium satisfies the equation $AG_0 = 1$, where the form of the differential operator A can be easily reconstructed. With the aid of this remark we derive from (4), taking (3) and (5) into account, the relation

$$(A\times 1 - 1\times A^*)\langle E\times E^*\rangle = \frac{4\pi\omega}{ic^2}(j\times\langle E^*\rangle + \langle E\rangle\times j^*) + [M\times 1 - 1\times M^* + (1\times\langle G^*\rangle - \langle G\rangle\times 1)K]\langle E\times E^*\rangle. \tag{A.1}$$

Performing the convolution (7) and using the optical theorem (6), we get

$$\int d\mathbf{r}\,\mathrm{div}\langle \Pi\rangle = -\frac{1}{2}\mathrm{Re}\int d\mathbf{r}\mathbf{j}\langle \mathbf{E}^*\rangle, \qquad \Pi = \frac{c}{8\pi}\mathrm{Re}[\mathbf{E}\times\mathbf{H}], \tag{A.2}$$

which is the integral form, averaged over the ensemble, of the electromagnetic-field energy-flux conservation law.

We replace now, purely arbitrarily, the bilinear combinations of the mean field $\langle E\rangle$ and the mean Green's tensor $\langle G\rangle$ by bilinear combinations of the incident field E_0 (2) and the Green's tensor G_0 (19) in a homogeneous medium:

$$\langle E\rangle\times\langle E^*\rangle \to E_0\times E_0, \quad \langle G\rangle\times\langle G^*\rangle \to G_0\times G_0^*. \tag{A.3}$$

A similar replacement is then effected in the right-hand side of (A.1) where, in addition, the first two terms in the square brackets vanish. We choose the intensity operator in the ladder approximation

$$K = \langle V\times V\rangle, \tag{A.4}$$

which is connected with the approximation (31) for the mass operator by the first-order optical theorem.[9] As a result we obtain in lieu of (A.2)

$$\int d\mathbf{r}\,\mathrm{div}\langle\mathbf{\Pi}\rangle = -\frac{1}{2}\,\mathrm{Re}\int d\mathbf{r}\mathbf{j}\mathbf{E}_0^* - \frac{c^2}{8\pi\omega}\iint d\mathbf{r}_1\,d\mathbf{r}_2\,(\mathrm{Im}\,M)_{\mu\nu}(\mathbf{r}_1,\mathbf{r}_2)\langle E_\nu(\mathbf{r}_2)E_\mu^*(\mathbf{r}_1)\rangle. \quad (A.5)$$

The second term in the right-hand side of this equation should be non-negative by virtue of (22). To demonstrate this more convincingly, we note that in the radiation-transport theory approximation[19] for Eq. (4), when

$$\langle E_\mu(\mathbf{r}_1)E_\nu^*(\mathbf{r}_2)\rangle = \int d^2n\,\exp(ik_0\mathbf{n}\mathbf{r})I_{\mu\nu}(\mathbf{R},\mathbf{n}), \quad \mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2, \quad \mathbf{r}=\mathbf{r}_1-\mathbf{r}_2, \quad (A.6)$$

where $I_{\mu\nu}(\mathbf{R},\mathbf{n})$ is the tensor of the radiant intensity at the point $\mathbf{R}$ and in the $\mathbf{n}$ direction, this term takes the form

$$\frac{c^2}{8\pi\omega}k_0h_1\iint d\mathbf{R}\,d^2nI_{\mu\mu}(\mathbf{R},\mathbf{n})>0. \quad (A.7)$$

Thus, the substitution (A.3) in Eq. (4) leads to violation of the electromagnetic-field energy-flux conservation law, with the scattering substance becoming energetically active.

We turn directly to derivation of Eq. (1). We represent the solution of (4) in the form

$$\langle E\times E^*\rangle = \langle E\rangle\times\langle E^*\rangle + \langle G\rangle\times\langle G^*\rangle\mathcal{U}\langle E\rangle\times\langle E^*\rangle, \quad (A.8)$$

where $\mathcal{U}$ is the sought operator. Substitution of (A.8) in (4) yields

$$\mathcal{U} = K + K\langle G\rangle\times\langle G^*\rangle\mathcal{U}. \quad (A.9)$$

We use, again purely arbitrarily, the substitution (A.3). It follows here from a comparison of (A.8) with (16) that the operator $\mathcal{U}$ coincides with U (9). We rewrite (A.9) in terms of the centroid coordinates and of a difference such as (A.6). We take the bilinear combination $G_0\times G_0^*$ in the Fraunhofer approximation[19]

$$G^0_{\mu\mu'}(\mathbf{r}_1-\mathbf{r}_1')G^{0*}_{\nu\nu'}(\mathbf{r}_2-\mathbf{r}_2') \approx P^{tr}_{\mu\mu'}(\mathbf{n}_{R-R'})\times P^{tr}_{\nu\nu'}(\mathbf{n}_{R-R'})\frac{1}{(4\pi|\mathbf{R}-\mathbf{R}'|)^2}\exp[ik_0\mathbf{n}_{R-R'}(\mathbf{r}-\mathbf{r}')], \quad (A.10)$$

where $\mathbf{n}_{R-R'}$ is a unit vector in the $\mathbf{R}-\mathbf{R}'$ direction. We denote by $L_0(\mathbf{R},\mathbf{n})$ the distance from the interior point $\mathbf{R}$ of the volume to its boundary in the $\mathbf{n}$ direction. We assume that when the product of this distance by a certain function of the variables $\mathbf{R}$ and $\mathbf{n}$ and over the volume of the substance and over all directions it is possible to take $L_0(\mathbf{R},\mathbf{n})$ outside the integral sign in the form of a certain mean value L_0. Under the above conditions we obtain from (A.9)

$$U_{\mu\mu',\nu\nu'}(\mathbf{n},\mathbf{n}_0) = \Omega K_{\mu\mu',\nu\nu'}(\mathbf{n},\mathbf{n}_0) + \frac{L_0}{(4\pi)^2}\int d^2n'K_{\mu\mu'',\nu\nu''}(\mathbf{n},\mathbf{n}')P^{tr}_{\mu''\mu'''}(\mathbf{n}')P^{tr}_{\nu''\nu'''}(\mathbf{n}') \times U_{\mu'''\mu',\nu'''\nu'}(\mathbf{n}',\mathbf{n}_0). \quad (A.11)$$

Here

$$U_{\mu\mu',\nu\nu'}(\mathbf{n},\mathbf{n}_0) = U_{\mu\mu',\nu\nu'}(k_0\mathbf{n},\,k_0\mathbf{n}_0;\,k_0\mathbf{n},\,k_0\mathbf{n}_0),$$

$$K_{\mu\mu',\nu\nu'}(\mathbf{n},\mathbf{n}') = \delta_{\mu\mu'}\delta_{\nu\nu'}\left(\frac{\omega}{c}\right)^4\left(\rho_0\frac{\partial\varepsilon_0}{\partial\rho_0}\right)_T^2 g_2(k_0(\mathbf{n}-\mathbf{n}')).$$

Equation (1) follows from (A.11) on the basis of (11), (15), and (33).

[1)] The symbols K and k_1 in Refs. 5 and 6 are the equivalents of out h and h_1.
[2)] The mean field (28) is transverse. We are therefore not considering the longitudinal electric-field component whose value is estimated in Ref. 11.
[3)] We note that according to (26) and (27) an opposite inequality of the form $h_1/k_0 \ll 1$, should also be satisfied.
[4)] According to Refs. 5 and 6, the characteristic dimension L_0 (1) for a sphere coincides with the radius of the latter.

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