

ON LIGHT SCATTERING BY DIFFUSE AUREOLE ABOUT AEROSOL PARTICLES HEATED BY LASER RADIATION

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It is shown that some methodical problems, responsible for speculations about aureole scattering of light, can be solved by using the asymptotic electrodynamic techniques.

1. PREFACE

The processes, occurring under the exposure of aerosol to high-power laser radiation (evaporation of liquid and solid particles, combustion of the latter, simple heating, i.e., the particles conversion into sources of heat in a gaseous medium, etc.), result in formation of aureoles, i.e. such areas around a particle, where the thermodynamic properties differ from those in the background atmosphere. In addition, such aureoles are certainly the optical inhomogeneities, influencing the light propagation. The detailed analysis of these problems can be found in monographs.¹⁻³

However, here arises rather a principal problem, whose essence is perfectly well outlined in Ref. 3. The consideration of aureole as an optically soft large particle is rather obvious, and means that

$$|m - 1| \ll 1, V \gg \lambda^3. \quad (1)$$

Here λ is the wavelength; m is the relative complex refraction index, V is the particle volume. Solution to the Maxwell equation for conditions (1) is well known, and some its details will be mentioned in Section 2.

However, if these results are used formally, the parameter

$$\eta = \frac{\text{coefficient of extinction by particle-aureole}}{\text{geometric cross section of aerosol particle}}$$

tends to infinity, as the aureole radius R increases, rather fast. (As follows from the estimates given in Ref. 3, R becomes equal to $\approx 100 \mu$ as early as 10^{-4} s, while the thermodynamic processes inherently require times of 10^{-3} – 10^{-2} s, for example $\eta \approx 200$ at $R \approx 350 \mu$. Analogous data on the anomalous growth of η are presented in Ref. 3 and references therein. Bukatyi and Kronberg recently have also obtained similar data.) Another important point is that the estimate $\eta = 0(\ln R)$ is a consequence of a seemingly correct solution.

At the same time, the experimental data, reviewed in Refs. 2 and 3, do not reveal similar growth of η . Some general ideas about such a drastic disagreement was proposed in Ref. 3, but they obviously are beyond the scope of electrodynamic.

The conviction that the problem, arising within the scope of electrodynamic, must be resolved just within this scope causes us to undertake the below analysis. It is quite realistic that the final result could be deduced merely from pure qualitative reasoning but we believe that the mathematical and methodical scrupulousness will not be excessive.

2. SOME RELATIONSHIPS FROM THE ELECTRODYNAMICS

The vector analog of the Kirchoff formula well-known (see, e. g., Ref. 4) in scalar optics is the expression⁵:

$$\mathbf{E}(\mathbf{r}) = -\frac{1}{4\pi} \int_{(\sigma)} d\sigma \{ (\mathbf{v} \text{ grad}' G) \mathbf{E}(\mathbf{r}') - \mathbf{v} (\mathbf{E}(\mathbf{r}') \text{ grad}' G) + G(\mathbf{v} \times \text{rot}' \mathbf{E}(\mathbf{r}')) + (\mathbf{E}(\mathbf{r}') \mathbf{v}) \text{ grad}' G \} \quad (2)$$

for the spectral component \mathbf{E} of the electric field strength at the point \mathbf{r} in vacuum. The Green's function $G(\mathbf{r}|\mathbf{r}') = |\mathbf{r} - \mathbf{r}'|^{-1} \exp(ik|\mathbf{r} - \mathbf{r}'|)$; $k = 2\pi/\lambda$; the closed surface σ with the normal \mathbf{v} to $d\sigma$ encloses \mathbf{r} ; $\mathbf{r}' \in \sigma$; grad' etc. designate the differentiation with respect to \mathbf{r}' . The system of coordinates (with the unit vectors $\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3$ and the x, y, z components of the vector \mathbf{r}) is shown in Fig 1, where σ consists of the plane $z = 0$ closed by the hemisphere with the radius $b \rightarrow \infty$; infinitely distant areas of σ certainly do not contribute into Eq. 2.

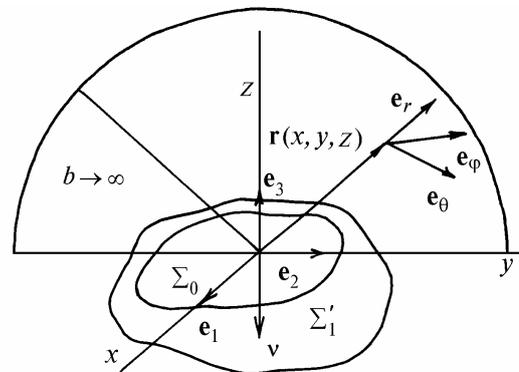


FIG. 1. The unit vectors $\mathbf{e}_r, \mathbf{e}_\theta,$ and \mathbf{e}_ϕ are set as in Fig. 2.

Let us consider now the field (hereinafter referred to as the external wave) of the form

$$\mathbf{E}_0(\mathbf{r}) = \mathbf{E}^{(0)}(x, y) \exp(ik \mathbf{k}_0 \mathbf{r}), \quad \mathbf{E}^{(0)} \mathbf{k}_0 = 0 \quad (3)$$

with the unit vector \mathbf{k}_0 of the Pointing vector. As to the function $\mathbf{E}^{(0)}(x, y)$, we believe, as usually, that it changes rather slowly as compared to \exp from Eq. (3). Actually, in our problem the sole purpose of using $\mathbf{E}^{(0)}$ is to underline the limitedness of a ray: $\mathbf{e}^{(0)} \neq 0$ within Σ_0 (see Fig. 1).

Substitution of Eq. (3) into Eq. (2) gives only condition $kr \equiv k |\mathbf{r}| \gg 1$, which is fulfilled almost automatically, and certainly the Maxwell equations for vacuum transform Eq. (3) into the expression

$$\mathbf{E}_0(\mathbf{r}) = - \frac{ik}{2\pi} \int_{(\Sigma')} ((\mathbf{e}_0 (\mathbf{e}_0 \mathbf{E}_0) + \mathbf{e}_\varphi (\mathbf{e}_\varphi \mathbf{E}_0)) G)_{z=0} dx dy \quad (4)$$

with the addition that $\Sigma' \ni \Sigma_0$ (see Fig. 1). A more detailed consideration of Eq. (4) one can find in Ref. 6.

Figure 2 illustrates the standard statement of the problem on scattering of the wave (3) by a particle with the volume V . The latter corresponds here to an aureole. For the internal ($\mathbf{r} \in V$) field $\mathbf{e}^{(j)}$ there is the integral equation^{4, 5, 7, 8}

$$\mathbf{E}^{(j)}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \text{rot rot} \int_{(V)} \frac{m-1}{2\pi} G(\mathbf{r}|\mathbf{r}') \mathbf{E}^{(j)}(\mathbf{r}') \quad (5)$$

equivalent to the Maxwell equations, and the first asymptotic of Eq. (1) is already included here. It happens so⁹ that the asymptotic of Eq. (1) allows one to neglect the influence of the initial electrodynamic conditions on the change of light polarization, i.e., the scalar description is applicable. The iterations of Eq. (5) can be therefore calculated using the effective approach from Ref. 10, with the following outcome:

$$\mathbf{E}^{(j)}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) \exp \left(ik \int_{-\infty}^z (m(x, y, z') - 1) dz' \right). \quad (6)$$

It should necessarily be emphasized that in Ref. 11 the result of the form (6), referred there to as the anomalous diffraction, is discussed only based on qualitative concepts. The above example demonstrates its derivation directly from the Maxwell equations. Let us also add that Eq. (5) was also used in Ref. 2.

The integral form of the Maxwell equations gives us a possibility to express the field \mathbf{E} outside the particle through $\mathbf{E}^{(j)}$. For the wave zone ($kr \gg 1$)

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \frac{k^2}{2\pi} \frac{\exp(ikr)}{r} \int_{(V)} d\mathbf{r}' (m-1) \{ \exp(-i(\mathbf{k}_0 \mathbf{r}')) \} \times \{ \mathbf{e}_0 (\mathbf{e}_0 \mathbf{E}^{(j)}(\mathbf{r}')) + \mathbf{e}_\varphi (\mathbf{e}_\varphi \mathbf{E}^{(j)}(\mathbf{r}')) \} \equiv \mathbf{E}_0 + \mathbf{E}_s \quad (7)$$

and \mathbf{E}_s in Eq. (7) is interpreted as the scattered wave. For the wave zone

$$G(\mathbf{r}|\mathbf{r}') \equiv (1/r) \exp(ikr - ik(\mathbf{r}' \mathbf{r}_0)) \quad (8)$$

with the designations from Fig. 2.

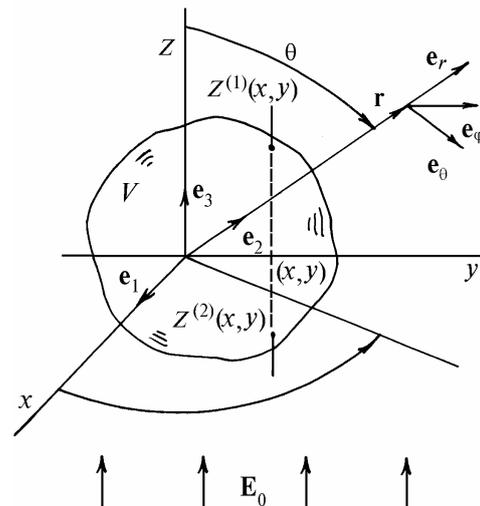


FIG. 2. In this figure, the following designation are used: \mathbf{r} is the point of observation of the scattered wave; V is the particle; \mathbf{e}_r , \mathbf{e}_θ , and \mathbf{e}_φ are the unit vectors of the spherical system for coordinates $\mathbf{r} = |\mathbf{r}|$, θ , and φ of the vector \mathbf{r} .

Remind also that just the form (7), describing the interference of external and scattered waves, implies the optical theorem, i.e. the expression for the extinction coefficient κ in terms of the amplitude (the factor before $(\exp ikr)/kr$ in \mathbf{E}_s from Eq. (7)) of the forward scattered wave ($\theta=0$). The function $\mathbf{E}_0(x, y)$ from Eq. (3) introduces some changes (overbar denotes the complex conjugation):

$$\kappa = \frac{2k}{|E_1^{(0)}(0)|^2 + |E_2^{(0)}(0)|^2} \text{Im} (A_1 \bar{E}_1^{(0)}(0) + A_2 \bar{E}_2^{(0)}(0))$$

with the values $A_\mu = \int d\mathbf{r}' (m-1) (\mathbf{e}_\mu \mathbf{E}^{(j)}) \exp ikz'$,

$\mu = 1$ and 2 . One can appreciate that κ can now prove to be formally dependent on Σ_0 (see. Fig. 1). And only for a plane wave, when $\mathbf{e}_0 = \text{const}$, $\kappa = 2k \text{Im} \int d\mathbf{r}' (m-1) (\exp ikz') \Phi$, where Φ is the factor after \mathbf{E}_0 in (6). Having substituted the explicit form of Φ we obtain

$$\kappa = 2 \operatorname{Re} \int dx' dy' (1 - \exp(ik \int_{-\infty}^{+\infty} (m-1) dz'))$$

automatically with the integration over V . The last equation gives, in particular, specific expressions which were used in Refs. 1–3 in the analysis of aureole scattering.

3. THE MAIN FEATURE OF THE AUREOLE SCATTERING

Substitution of Eq. (6) into Eq. (7) and some simple transformations (again following Ref. 10) yield

$$\begin{aligned} \mathbf{E}_s &= \mathbf{e}_0 (\mathbf{e}_0 \mathbf{C}) + \mathbf{e}_\varphi (\mathbf{e}_\varphi \mathbf{C}); \\ \mathbf{C} &= (1/r) (\exp ikr) \frac{ik}{2\pi} \int dx' dy' (\exp(iq_1 x' + iq_2 y')) \times \\ &\times \mathbf{E}^{(0)}(x', y') \left\{ 1 - \exp\left(ik \int_{-\infty}^{+\infty} (m(x', y', z') - 1) dz'\right) \right\} \end{aligned} \quad (9)$$

with the projections $\mathbf{q} = \mathbf{k}_0 - \mathbf{r}_0$ onto the axes \mathbf{e}_1 and \mathbf{e}_2 . Additional simplification is due to the fact that the asymptotic (1) allows neglecting the terms related to $q_3 = (\mathbf{q}\mathbf{e}_3) = 1 - \cos\theta$. (The physical reason for this is the strongly forward peaked scattering phase function.)

Let us now consider the versions presented schematically in Fig. 3. The structure of the integral

(9) is quite clear: $\int_{-\infty}^{+\infty} dz'(\dots)$ is, in fact, $\int_{z_1(x', y')}^{z_2(x', y')} dz'(\dots)$

with z_1 and z_2 from Fig. 2, and then the integration in Eq. (9) is performed over the area in the plane $z = 0$. For the case shown in Fig. 3a, it will be the projection of “a particle” onto the plane $z = 0$, the integration variables form V , and the first term in $\{\dots\}$ in Eq. (9) is the usual description of the Fraunhofer diffraction. The situation changes drastically for the case (b): now

$\int dx' dy'$ is the integration over the beam cross section,

and relationships (4), (8), and (9) transform the first term in Eq. (9) into $(-\mathbf{E}_0)$ and now it and the first term of Eq. (7) cancel each other. Now the field outside the particle is described by the vector

$$\begin{aligned} \mathbf{B}(\mathbf{r}) &= \frac{\exp(ikr)}{r} \frac{ik}{2\pi} \int_{(\Sigma_0)} dx' dy' \mathbf{E}_0(x', y') \times \\ &\times \exp\left(ik \int_{z_1}^{z_2} (m-1) dz' - ik(r_{01}x' + r_{02}y')\right). \end{aligned} \quad (10)$$

In Eq. (10) the property of \mathbf{k}_0 from Eq. (3) is taken into account as well as the designations from Figs. 1 and 2.

The last circumstance certainly changes the interpretation. There is no diffraction on the particle contour and the suspicion arises, which however merges into a belief, that only the geometric optics rays remain. This fact will be demonstrated, and quite clearly, later on. Now we would like only to note that, as it follows from the remark on the extinction coefficient in the end of Section 2, with the disappearance of \mathbf{E}_0 from Eq. (7) the optical theorem also disappears together with the formal consequences for κ .

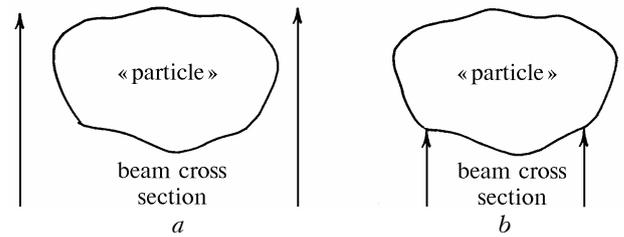


FIG. 3. Classification of situations for Eq. (9).

We shall make a small retrospective journey into the problem on asymptotic (with the main condition $V \gg \lambda^3$) solution of the Maxwell equations before explaining the methodical meaning of Eq. (10). Under the assumption that inside V the geometric optics is valid, for the field outside V (we already consider the case shown in Fig. 3b) the integral form (2) yields the following expression

$$\begin{aligned} \mathbf{E}(\mathbf{r}) &= \frac{k^2}{4\pi} \int dx' dy' \int_{z_1(x', y')}^{z_2(x', y')} dz' (m^2 - 1) \times \\ &\times \mathbf{E}(\mathbf{r}') (\exp ikS(\mathbf{r}')) G(\mathbf{r}|\mathbf{r}'). \end{aligned} \quad (11)$$

The eikonal S and the “slow” (as compared to $\exp(ikS)$) factor \mathbf{E} enter into this expression. As usually, $\int dz'$ can be treated using the integral of the

type $J = \int_{z_1(x', y')}^{z_2(x', y')} dz' g(\xi) \exp(ikh(\xi))$ with the formal

condition that $k \rightarrow \infty$. If the equation $h'(\xi) = 0$ has no roots on the integration interval, then the result of J estimation as the integral of the Fourier type will be the following¹²:

$$J = \frac{g(z_2)}{ikh'(z_2)} \exp(ikh(z_2)) - \frac{g(z_1)}{ikh'(z_1)} \exp(ikh(z_1)).$$

In Eq. (11) $S + |\mathbf{r} - \mathbf{r}'|$ plays the role of h , and, as follows from the eikonal equation, $h'(\xi) = 0$ is equivalent to $mt_3 - R_{03} = 0$. Here t_3 and R_{03} are the components, along the \mathbf{e}_3 unit vector, of the vectors \mathbf{t} , normal to the geometric optics ray trajectory, and $\mathbf{R}_0 = (\mathbf{r} - \mathbf{r}') / |\mathbf{r} - \mathbf{r}'|$.

The first fine feature is connected with the statement that the internal points of V cannot be the roots of the last equation; this, methodical, in its essence, problem is considered in Appendix (see below).

The other feature is due to the vanishing of g on the surface, since $m \rightarrow 1$ there. But if at the surface points $h' = 0$ also because of $m \rightarrow 1$, then once the uncertainty is removed, the estimate remains the same. The obvious condition of the latter is $t_3 = R_{03}$; by a proper choice of the coordinate system it is easy to reach $\mathbf{t} = \mathbf{R}_0$ with the clear physical meaning: the field at the point \mathbf{r} is generated by the geometric optics ray, which is directed immediately from the surface V into \mathbf{r} . It is clear that in the problem considered the points $z_2(x', y')$ are needed.

One more point to be noted is the construction of \mathbf{E} . For Eq. (3) and as follows from the geometric optics, one should write $\mathbf{E} = \mathbf{E}^{(0)} e.p(ikz_1)$, that is, the field at the "entrance" (for the external wave) side of V .

If one will follow the above scenario of the asymptotic estimate and use again Eq. (7) then

$$\text{Eq. (11) will transform into Eq.(10) at } S = \int_{-\infty}^z m dz'$$

with the clear $(-\infty)$ taken as the lower limit of integration. And it only remains for us to ensure that such a value is truly treated as eikonal.

The formal aspect of the problem is obvious – the values $(1/m) \text{grad} S$, $\text{rot}((1/m) \text{grad} S)$, $\rho(l) = \mathbf{t}$ are necessarily to be agreed: the former should give the unit vector \mathbf{t} , which will determine the ray trajectory ρ as a function of its length l ; the vanishing of the latter will be indicative of the existence of nonintersecting elementary rays. In calculations with the S , written, and for the rather acceptable assumption about the aureole properties as microscopic (practically the constant characteristics at distances of the order of λ), $\ln m \equiv \ln(1 + \beta) \cong \beta$ will appear, and $|\beta|$ is the obvious parameter of smallness in the asymptotic (1). Under such an assumption, the unit vector \mathbf{e}_3 serves as \mathbf{t} , and the addition to it is $O(\beta)$, that is in a good agreement with the geometric optics of "soft" media^{11,13} with its practically straight line rays; $|\text{rot}(1/m) \text{grad} S| = O(\beta^2)$.

4. DIFFUSE AUREOLE IN THE TRANSFER EQUATION

It is certainly evident that the particle embedded in aureole is treated as an actually scattering particle with all its diffraction features (scattering phase function, optical theorem, etc.) only with a small aureole (Fig. 3a). The consequence is also clear: the

previous "aerosol" form of the transfer equation only with the characteristics of "compound" particles.

Quite different pattern arises with large aureole. The previous analysis, in effect, makes it clear that for the situation shown in Fig. 3b to take place it is sufficient that the aureole size is greater than the width of elementary geometric optics ray in the external, with respect to aerosol particle, medium. Of course, the situation becomes more certain, when the aureoles from different particles overlap each other or each of them is greater than the coherence length of the amplitude $\mathbf{E}^{(0)}(x, y)$; meanwhile in the latter case it would be interesting to follow experimentally the qualitative jump between the cases shown in Figs. 3a and b.

Section 3 concerns the problem on interaction of the field with the system "aerosol particles + aureoles." (Some its fine details are discussed in Ref. 2.) The exact relation (of the form (5) or (7)) between the external, \mathbf{e} , and internal, $\mathbf{e}^{(j)}$, fields via the integral over the volume of optical inhomogeneity, immediately leads us to the expression $\mathbf{e} = \mathbf{e}_1 + \mathbf{e}_2$, where \mathbf{e}_1 and \mathbf{e}_2 are the results of integration over the area of a particle and an aureole.

The statement of the exact problem for $\mathbf{e}^{(j)}$ with the boundary conditions of electrodynamics on the interface between media with different complex refractive indexes is indicative of the necessity to introduce some corrections into $\mathbf{e}^{(j)}$, which would occur if the external wave would fall directly upon the aerosol particle. However, some efficient simplifications are rather clear here.

Naturally, the possibility of describing the field inside the aureole according to the rules of geometric optics and the asymptotic (1) allow us to state that the field $\mathbf{e}^{(j)}$ for the particle formally remains the same (as without aureole), but the relative complex refractive index of the particle should be replaced by $m^{(j)} / (m^{(0)} + \delta m^{(0)})$; here $m^{(j)}$ and $m^{(0)}$ are the complex refractive indexes of the particulate matter and the external (not perturbed by aureole) atmosphere; $\delta m^{(0)}$ is change of $m^{(0)}$ due to aureole of the particle at its side exposed to the external wave. It is hardly probable that such a substitution affects markedly the optical characteristics of the aerosol particle.

Then, the further processes the light scattered by the particle is involved in are governed, as early, by the geometric optics and therefore it is sufficient to introduce the absorption by "aureole medium" and refraction in it into the transfer equation. Properties of the aureole medium itself are determined by the problem on medium heating by point sources distributed over it with the subsequent averaging over the intervals of their statistical parameters.

The reasoning presented above is summarized in the equation for the light intensity $I(\mathbf{r}, \mathbf{n})$ at the point \mathbf{r} and in the direction of the unit vector \mathbf{n} :

$$\mathbf{n} \text{ grad } I = -(\chi + \alpha) I + (\text{multiple scattering by aerosols}) \tag{12}$$

Here χ designates the sum of aerosol extinction coefficient and the molecular atmospheric absorption; α is the absorption coefficient of the aureole medium; and for \mathbf{n} we can write ordinary equation of refraction in the same aureole medium.

Strictly speaking, as it follows from the physical meaning of the transfer equation, I is the intensity of the elementary geometric optics ray, what emphasizes once more the necessity to turn to the situation shown in Fig. 3b. It is clear that when solving Eq. (12), that anomalously large extinction of I , which was discussed in Section 1, will appear in no way.

The problem of α in Eq. (12) is independent: the value of this coefficient can be calculated directly for the aerosol substance evaporated or through the change in the refraction index, that can be recalculated into α by the dispersion relationships.

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APPENDIX

Under the assumption of geometric optics (with formal $k \rightarrow \infty$) the Poynting vector is $\mathbf{\Pi} = 2c (\exp(-2k \operatorname{Im} S)) (\mathbf{E} \mathbf{E}) (\operatorname{Re} m \mathbf{n})$, where c is the speed of light and $\mathbf{E} = \mathbf{E} \exp(ikS)$ was already discussed. We now consider the complex $m = m' + im''$, and from the equation of eikonal $\operatorname{grad} S = m \mathbf{n}$ the complex character of $\mathbf{n} = \mathbf{n}' + i\mathbf{n}''$ follows. With the same notes, $\operatorname{div} \mathbf{\Pi} = -2kc (\mathbf{E} \mathbf{E}) (\exp(-2k \operatorname{Im} S)) (\operatorname{Re} m \mathbf{n}) (\operatorname{Im} m \mathbf{n}) = Q$, i.e. the amount of heat releasing in a unit volume. The definition of Q via \mathbf{e} and the dipole moment leads, as early within the scope of geometric optics, to the expression $Q = kc (\mathbf{E} \mathbf{E}) (\operatorname{Im} \epsilon) (\exp(-2k \operatorname{Im} S))$, where the permittivity $\epsilon = m^2$. The equations written fit the equalities $\operatorname{Re} m \mathbf{n} \operatorname{Im} m \mathbf{n} = m' m'' = \epsilon''/2$, which commonly are derived from the above listed complex parameters and the condition $\mathbf{n}^2 = 1$.

We come back now to the equation $m n_3 - R_{03} = 0$ and assume that its root is the point $\mathbf{r}_0 \in V$, i.e. the domain, where the above expressions hold true. In our general case, we impose no conditions upon the choice of coordinate axes, therefore the third axis can

be believed coincident with the direction $\mathbf{\Pi}$ at the point \mathbf{r}_0 . Because R_{03} is real and as follows from the equation for $\mathbf{\Pi}$, the projection of $\operatorname{Im} m \mathbf{n}$ on $\operatorname{Re} m \mathbf{n}$ equals zero, i.e. in fact $\operatorname{div} \mathbf{\Pi} = 0$. However, $Q \neq 0$, although absorption, though weak, always exists,¹⁴ and moreover the second definition of Q says about its independence of the direction of wave propagation.

Just the contradiction obtained ($\operatorname{div} \mathbf{\Pi} = Q$) proves the statement about the roots of equation $h'(\xi) = 0$ which was used in Section 3.

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