Method of integral equations and an extinction theorem in bulk and surface phenomena in nonlinear optics

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This work is a response to a problem which was most clearly formulated by Wolf [*Coherence and Quantum Optics,* edited by L. Mandel and E. Wolf (Plenum, New York, 1973), p. 339] in this way: "Attempts to generalize [the extinction theorem] within the framework of molecular optics encounters formidable difficulties." Here the method of integral equations is applied to an arbitrary nonlinear and anisotropic medium, taking into account quadrupole and magnetic-dipole radiation. Using the fundamental equations of molecular optics, we prove the extinction theorem in a general case, and its physical interpretation is elucidated. The question about structure of a surface layer that produces the reflected wave is clarified. A connection between the microscopic and macroscopic characteristics of nonlinear media is obtained. This advance was achieved by the implementation of the straightforward idea of variable substitution in the original integral equation. This substitution turns out to yield insight into the transition from a local to a Maxwellian field.

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I. INTRODUCTION

The method of integral equations (MIE) in molecular optics [1] is based on the representation of the medium as a system of discrete oscillators. In contrast to the more usual spatial-averaging approach of Maxwell's equations, the MIE starts from the discrete model of a medium and gives a self-consistent description of electromagnetic phenomena in the framework of such a model. In the case of linear optics, the method of integral equations turns out to be useful when one considers problems such as spatial dispersion [2,3], the scattering of the light by ultrasound [4], and the propagation of light through layered media [5].

What is the reason for using the MIE approach for consideration of optical problems? Optics was "born in a vacuum," but already its first steps into a medium stimulated the question: "What is a medium from the viewpoint of light"? In other words, is the medium a continuous substance without any interval structure, or does it consist of discrete elementary radiators? The Maxwell differential-equations approach is based on the first (a continuous substance) premise, but it is known that in reality matter is composed of discrete elementary radiators. The problem of reconciling both these approaches was formulated by Esmarch, Oseen, and Ewald in the years 1912–16, and was solved for linear isotropic homogeneous media (for a modern review see Ref. [1]). The main result may be stated as the Ewald-Oseen extinction theorem (EOET) which shows how, due to the interference of the separate elementary oscillators, an incident wave from the vacuum is canceled in the medium and a new wave propagating in the medium with a reduced velocity appears. Originally, EOET was proved only for a linear, electric-dipole homogeneous isotropic media. Several generalizations of this theorem have been proved for electric-quadrupole and magnetic-dipole linear, isotropic, semi-infinite media on which a plane wave is incident [6], and for nonlinear media under very special conditions: The medium must be semi-infinite, homogeneous, and isotropic, and must produce a plane wave of nonlinear polarization. In addition, a preset field approximation is valid [7]. As will be seen later, such assumptions are not accidental, and in the approaches of Refs. [6,7] the results could not have been obtained without them.

The important result of these investigations was the demonstration of the possibility of matching the microscopic approach to the Maxwell equations for nonlinear and linear quadrupole media, at least in some specific situations. However, without these special assumptions the proof of EOET is not only destroyed, but also it seems that, in a general case, the electromagnetic field cannot satisfy the integral and wave equations simultaneously. Thus the problem is such that it requires consideration from a general point of view.

A completely different approach to the extinction theorem was developed by Sein, who demonstrated that the extinction theorem is a natural consequence of Maxwell equations and material equations in the simple situation of no charge and current singularities [8]. Later on, generalizations of this approach to the singular charge and current densities in the medium were made [9,10]. Therefore, the EOET, in principle, does not contradict the Maxwell equations.

However, these investigations do not touch on the subject of microscopic analysis. As a result, the question of generalizing the EOET within the framework of molecular optics remains open. The results of Refs. [6,7] stimu-

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late additional interest in the general problem because from them it follows that, with the exception of some special cases, there is no obvious way to solve it. Interest in this question is doubled when one realizes that in the general case an electromagnetic field cannot satisfy integral and wave equations simultaneously.

For the solution of this problem we adopt here the principal idea of a formal "substitution of variables" in the integral equation in such a form that the new variables satisfy wave and integral equations simultaneously [11,12]. We accomplish this program for the most general case of nonlinear anisotropic inhomogeneous media with allowance made for not only the electric-dipole but the electric-quadrupole and magnetic-dipole moments of the elementary oscillators as well. It turns out that a consequence of properly taking into account the quadrupole radiation, apart from the generalization of the EOET, is that it reveals an actual aspect for the understanding of the extinction theorem's physical meaning [12].

In Sec. II the problem of matching the integral-equation approach and the Maxwell-equation approach is considered, and an extinction theorem for a medium with allowance for quadrupole and magnetic-dipole mechanisms of nonlinearity is deduced. In Secs. III and IV the reflected-wave and local-field corrections are discussed. The connection between our approach and those considered previously is given in Sec. V. The main results are listed in Sec. VI.

II. THE PROBLEM OF MATCHING THE MIE APPROACH AND MAXWELL-EQUATION APPROACH AND AN EXTINCTION THEOREM

First we define the contributions of the medium to the fields. These include electric-dipole, quadrupole, and magnetic-dipole terms. The expressions for the microscopic fields $\mathbf{E}_d$, $\mathbf{H}_d$ of the electric dipole, $\mathbf{E}_q$, $\mathbf{H}_q$ of the quadrupole, and $\mathbf{E}_m$, $\mathbf{H}_m$ of the magnetic dipole are

$$\mathbf{E}_d(\mathbf{r},t)=\nabla\times\nabla\times\frac{\mathbf{d}\left[\mathbf{r}',t-\frac{R}{c}\right]}{R}, \tag{1a}$$

$$\mathbf{H}_d(\mathbf{r},t)=\frac{1}{c}\nabla\times\frac{\dot{\mathbf{d}}\left[\mathbf{r}',t-\frac{R}{c}\right]}{R}, \tag{1b}$$

$$\mathbf{E}_q(\mathbf{r},t)=-\nabla\times\nabla\times\nabla\cdot\frac{\hat{q}\left[\mathbf{r}',t-\frac{R}{c}\right]}{R}, \tag{2a}$$

$$\mathbf{H}_q(\mathbf{r},t)=-\frac{1}{c}\nabla\times\nabla\cdot\frac{\dot{\hat{q}}\left[\mathbf{r}',t-\frac{R}{c}\right]}{R}, \tag{2b}$$

$$\mathbf{E}_m(\mathbf{r},t)=-\frac{1}{c}\nabla\times\frac{\dot{\mathbf{m}}\left[\mathbf{r}',t-\frac{R}{c}\right]}{R}, \tag{3a}$$

$$\mathbf{H}_m(\mathbf{r},t)=\nabla\times\nabla\times\frac{\mathbf{m}\left[\mathbf{r}',t-\frac{R}{c}\right]}{R}, \tag{3b}$$

where $\mathbf{R}=\mathbf{r}-\mathbf{r}'$. Here $\mathbf{r}$ is the coordinate of the observation point; $\mathbf{r}'$ is the coordinate of the radiator; and $\mathbf{d}(\mathbf{r}',t)$, $\hat{q}(\mathbf{r}',t)$, and $\mathbf{m}(\mathbf{r}',t)$ are the electric-dipole, quadrupolar, and magnetic-dipole moments, respectively. The gradient symbol ∇ indicates differentiation over $\mathbf{r}$. Time derivatives are indicated by a dot above the quantity differentiated. A dot at midline indicates the contraction of two tensors over one pair of indices. Note that Eqs. (1)–(3) yield well-known forms of the fields in the various radiation zones. When carrying out the spatial differentiation operations, the number of such operations either in the numerator or denominator determines the power law of the field decrease with distance, i.e., the type of zone (static, wave, or intermediate). From formula (2) it is evident that we can add to $\hat{q}$ a unit tensor multiplied by an arbitrary scalar function without changing the fields. This means we can always choose a tensor $\hat{q}$ such that Tr $\hat{q}=0$. This degree of freedom in $\hat{q}$ was previously demonstrated only in the far-distant zone or for a static field. Also, it is easy to demonstrate that $\hat{q}$ is a symmetric tensor, since its antisymmetric part can be removed by means of renormalization of the magnetic moment.

Let us consider Fourier transforms of the fields $\mathbf{E}'(\hat{r}_l),\mathbf{H}'(\hat{r}_l)$ at the frequency ω. We place a radiating atom at the point $\mathbf{r}_l$, and find

$$\mathbf{E}'(\mathbf{r}_l)=\mathbf{E}_i(\mathbf{r}_l)+\sum_{j(\neq l)}[\nabla\times\nabla\times\mathbf{d}(\mathbf{r}_j)G(R_{jl}) -\nabla\times\nabla\times\nabla\cdot\hat{q}(\mathbf{r}_j)G(R_{je}) +ik\nabla\times\mathbf{m}(\mathbf{r}_j)G(R_{jl})], \tag{4a}$$

$$\mathbf{H}'(\mathbf{r}_l)=\mathbf{H}_i(\mathbf{r}_l)+\sum_{j(\neq l)}[\nabla\times\nabla\times\mathbf{m}(\mathbf{r}_j)G(R_{jl}) +ik\nabla\times\nabla\cdot\hat{q}(\mathbf{r}_j)G(R_{jl}) -ik\nabla\times\mathbf{d}(\mathbf{r}_j)G(R_{jl})], \tag{4b}$$

$$G(R_{jl})\equiv\frac{e^{ikR_{jl}}}{R_{jl}},\quad R_{jl}\equiv|\mathbf{r}_j-\mathbf{r}_l|,\quad k=\frac{\omega}{c},$$

where $\mathbf{E}_i$ and $\mathbf{H}_i$ are the strengths of the electric and magnetic fields of the incident waves.

For given values of $\mathbf{d}$, $\hat{q}$, and $\mathbf{m}$, formulas (4) determine the fields at $\mathbf{r}_l$ due to all other radiators, except for the radiator l, and also due to the incident wave. The radiation from a source depends on the fields $\mathbf{E}'(r_j),\mathbf{H}'(r_j)$ [this dependence is determined by $\mathbf{d}=\mathbf{d}(\mathbf{E}',\mathbf{H}'),\hat{q}=\hat{q}(\mathbf{E}',\mathbf{H}'),\mathbf{m}=\mathbf{m}(\mathbf{E}',\mathbf{H}')$ of each radiator] and so formulas (4) are actually equations for the fields $\mathbf{E}'$ and $\mathbf{H}'$, which must be solved self-consistently. Our task is to analyze the characteristics of the solutions of these microscopic equations and to compare them with the solutions of the macroscopic Maxwell equations.

As is commonly done in linear optics, let us pass from the summation to the integration and isolate the contribution from the radiators inside a Lorentz sphere σ, with its center at the point $\mathbf{r}_l=\mathbf{r}$. We choose the radius a of the sphere to be large compared to the distance b between the radiators, but small compared to the scale of the incident inhomogeneous field and the scale of the spatial distribution of the radiators. As a result, we obtain the following integral equations for $\mathbf{E}'$ and $\mathbf{H}'$:

$$\mathbf{E}'(\mathbf{r}')=\mathbf{E}_i(\mathbf{r})+\mathbf{E}_\sigma(\mathbf{r}) + \int_\sigma^\Sigma (\nabla\times\nabla\times\mathbf{P}G-\nabla\times\nabla\times\nabla\cdot\hat{Q}G + ik\nabla\times\mathbf{M}G)d^3\mathbf{r}' , \quad (5a)$$

$$\mathbf{H}'(\mathbf{r}')=\mathbf{H}_i(\mathbf{r}')+\mathbf{H}_\sigma(\mathbf{r}') + \int_\sigma^\Sigma (-ik\nabla\times\mathbf{P}G+ik\nabla\times\nabla\cdot\hat{Q}G + \nabla\times\nabla\times\mathbf{M}G)d^3\mathbf{r}' , \quad (5b)$$

where Σ is the boundary of the medium, and $\mathbf{P}$, $\mathbf{M}$, and $\hat{Q}$ are the electric-dipole, magnetic-dipole, and quadrupole volume densities, respectively.

Unlike a traditional approach, in which the field acting on the oscillators inside a Lorentz sphere is considered to be uniform, here we take into account the change of field in first order. To determine $\mathbf{E}_\sigma$ and $\mathbf{H}_\sigma$, we consider all dipoles inside the sphere σ to be identical. Then it is obvious from Eqs. (1) that the contribution of these dipoles to $\mathbf{E}_\sigma$ is $\hat{\gamma}\cdot(\mathbf{d}/b^3)\equiv\hat{\gamma}\cdot\mathbf{P}$, where b^3 is the volume per radiator, and $\hat{\gamma}$ is a dimensionless second-rank tensor, which is determined by the spatial distribution of the radiators (for example, the geometry of the crystalline lattice):

$$(\hat{\gamma})_{st}=b^3\sum_j^\sigma \frac{3(\mathbf{n}_{jl})_s(\mathbf{n}_{jl})_t-\delta_{st}}{R_{jl}^3} , \quad \mathbf{n}_{jl}\equiv\frac{\mathbf{R}_{jl}}{R_{jl}}\equiv\frac{\mathbf{r}_j-\mathbf{r}_l}{|\mathbf{r}_j-\mathbf{r}_l|} . \quad (6a)$$

Here indices s and t label the Cartesian components, and δ_{st} is the Kroneker symbol. Using a first-order approximation to the spatial dispersion of $\mathbf{d}$ inside σ, we obtain the term $\hat{\gamma}_1:\nabla\mathbf{d}/b^2\equiv b\hat{\gamma}_1:\nabla\mathbf{P}$, where $\hat{\gamma}_1$ is the third-rank tensor:

$$(\hat{\gamma}_1)_{stp}=b^2\sum_j^\sigma \frac{3(\mathbf{n}_{jl})_s(\mathbf{n}_{jl})_t(\mathbf{n}_{jl})_p-\delta_{st}(\mathbf{n}_{jl})_p}{R_{jl}^2} . \quad (6b)$$

By analogy, contributions of the quadrupole and magnetic-dipole moments may be written in the following:

$$\mathbf{E}_\sigma(\mathbf{r})=\hat{\gamma}\cdot\mathbf{P}(\mathbf{r})+\frac{1}{b}\hat{\xi}:\hat{Q}(\mathbf{r})+ikb\hat{\gamma}_M\cdot\mathbf{M}(\mathbf{r}) + b\hat{\gamma}_1:(\nabla\mathbf{P})+\hat{\xi}_1:(\nabla\hat{Q}) , \quad (7a)$$

$$\mathbf{H}_\sigma(\mathbf{r})=\hat{\gamma}\cdot\mathbf{M}(\mathbf{r})+ik\hat{\xi}_M:\hat{Q}(\mathbf{r})-ikb\hat{\gamma}_M\cdot\mathbf{P}(\mathbf{r}) + b\hat{\gamma}_1:(\nabla\mathbf{M}) . \quad (7b)$$

It follows from Eqs. (1)–(3) that formulas for $\hat{\xi}$, $\hat{\xi}_1$, $\hat{\gamma}_M$, $\hat{\xi}_M$ take a form

$$(\hat{\xi})_{stp}=-b^4\sum_j^\sigma \frac{15(\mathbf{n}_{jl})_s(\mathbf{n}_{jl})_t(\mathbf{n}_{jl})_p-6\delta_{st}(\mathbf{n}_{jl})_p}{R_{jl}^4} , \quad (8a)$$

$$(\hat{\xi}_1)_{stpq}=-b^3\sum_j^\sigma \frac{15(\mathbf{n}_{jl})_s(\mathbf{n}_{jl})_t(\mathbf{n}_{jl})_p(\mathbf{n}_{jl})_q-6\delta_{st}(\mathbf{n}_{jl})_p(\mathbf{n}_{jl})_q}{R_{jl}^3} , \quad (8b)$$

$$(\hat{\gamma}_M)_{st}=b^2\sum_j^\sigma (\hat{\epsilon})_{sqt}\frac{(\mathbf{n}_{jl})_q}{R_{jl}^2} , \quad (8c)$$

$$(\xi_M)_{stp}=3b^3\sum_j^\sigma (\hat{\epsilon})_{sqp}\frac{(\mathbf{n}_{jl})_q(\mathbf{n}_{jl})_t}{R_{jl}^3} . \quad (8d)$$

Here $\hat{\epsilon}$ is an antisymmetric unit tensor of the third rank; and indices p and q, as well as s and t number the Cartesian components. In Eqs. (8c) and (8d) summation over the index "q" is implied.

In the general case the components of the tensors $\hat{\gamma}$ and $\hat{\xi}$ are of the order of unity. For a medium of randomly distributed radiators we have (see Appendix A)

$$\hat{\gamma}=\hat{\gamma}_1=\hat{\gamma}_M=\hat{\xi}=\hat{\xi}_1=\hat{\xi}_M=0 . \quad (9)$$

Equations (7) actually include all the possible versions of the construction of the vectors $\mathbf{E}_\sigma$ and $\mathbf{H}_\sigma$ from the vectors ∇, $\mathbf{P}$, $\mathbf{M}$, and the tensor $\hat{Q}$. Here we took into account that the magnetic field of the electric dipole and electric field of the magnetic dipole inside the Lorentz sphere are small in the parameter kb [terms $\mathbf{P}(\mathbf{r})$ in Eq. (7b) and $\mathbf{M}(\mathbf{r}')$ in Eq. (7a)]. In connection with the inclusion of the spatial dispersion effects, i.e., terms with the operator ∇ in Eq. (7), the tensors $\hat{\gamma}$ and $\hat{\xi}$ must not be taken in a static-field approximation, as commonly used, but with allowance for the terms $kR_{jl}\sim ka$ in Eq. (1), where a is the Lorentz sphere radius. Because of the structure of Eqs. (1), the factor $(1-ikR_{jl})e^{ikR_{jl}}$ in Eqs. (6) and (8) appears. But to an accuracy of quadratic terms, this factor equals unity. Therefore, within the accuracy up to terms of the order of ikR_{jl}, inclusive, Eqs. (6) and (8) remain the same as in the static-field approximation.

Due to the random distribution of the oscillators, any internal shell inside the Lorentz sphere makes a zero contribution to tensors $\hat{\gamma}$ and $\hat{\xi}$. For a periodic distribution of the oscillators a contribution from the periphery of the Lorentz sphere will be nearly the same as that for a chaotic medium, and so is zero as well. This effect is due to the specific universal angular dependence of Eqs. (1) of the Lorentz sphere's periphery under arbitrary, random or periodical distribution of the oscillators. This result removes the problem of a logarithmic divergence at the upper limit of the summation in Eqs. (6) and (8).

A natural assumption (see Ref. [1]) that the solutions of the integral equations (5) for $\mathbf{E}'$ and $\mathbf{H}'$ must satisfy the wave equation with a certain velocity of propagation, and that induced forces are proportional to $\mathbf{P}^{\mathrm{NL}}$, $\hat{Q}$ and $\mathbf{M}^{\mathrm{NL}}$ (where $\mathbf{P}^{\mathrm{NL}}$ and $\mathbf{M}^{\mathrm{NL}}$ are the $\mathbf{E}'$- and $\mathbf{H}'$-dependent nonlinear parts of $\mathbf{P}$ and $\mathbf{M}$) contradicts, in a general case, the original integral equations (5). This contradiction is not surprising as there is no reason to believe that the *microscopic* fields $\mathbf{E}'$ and $\mathbf{H}'$ in the medium will satisfy the *macroscopic* wave equations as well. *We will see that in reality in most cases it is impossible to construct the wave equation in such a form.* On the other hand, in the model of a really spatially homogeneous medium the wave equations for electric and magnetic fields do exist. It is reasonable to assume that in the model of a medium made up from discrete oscillators these wave equations must correspond to the wave equations for macroscopic (averaged) fields. The well-known connection between microfields and macrofields for isotropic electric-dipole media, as well as Eq. (7), suggest the idea of substitution of the variables in the equation (5) in the form

$$\mathbf{E}=\mathbf{E}'+\hat{\beta}\cdot\mathbf{P}+\frac{1}{b}\hat{\eta}:\hat{Q}+ik\hat{\beta}_M\cdot\mathbf{M}+b\hat{\beta}_1:\nabla\mathbf{P}+\hat{\eta}_1\vdots(\nabla\hat{Q}) , \tag{10a}$$

$$\mathbf{H}=\mathbf{H}'+\hat{\beta}\cdot\mathbf{M}+ik\hat{\eta}_M:\hat{Q}-ik\hat{\beta}_M\cdot\mathbf{P}+b\hat{\beta}_1:\nabla\mathbf{M} . \tag{10b}$$

Here $\hat{\beta}$ and $\hat{\eta}$ are free parameters. Their values are chosen in such a manner that the fields $\mathbf{E}$ and $\mathbf{H}$ would satisfy certain wave equations as well. We would like to emphasize that all the above-mentioned considerations are not obligatory, and one may come to substitution (10) starting from a formal mathematical condition that the new variables $\mathbf{E}$ and $\mathbf{H}$ must satisfy both the integral equations and the wave equations.

Let us substitute (10) into (5), and take into account that

$$\nabla\times\nabla\times\nabla\times\mathbf{P}G=k^2\nabla\times\mathbf{P}G , \tag{11a}$$

$$\nabla\times\nabla\times\nabla\times\mathbf{M}G=k^2\nabla\times\mathbf{M}G , \tag{11b}$$

$$\nabla\times\nabla\times\nabla\times\nabla\cdot\hat{Q}G=k^2\nabla\times\nabla\cdot\hat{Q}G . \tag{11c}$$

We may factor the operator $\nabla\times\nabla\times$ outside the integral sign. Since the boundary of integration includes the surface of the sphere with the center at the point $\mathbf{r}$, then operations of differentiation over $\mathbf{r}$ and integration over $\mathbf{r}'$ do not commute with each other (see Appendix B). Suppose that $\mathbf{E}$ and $\mathbf{H}$ satisfy, in addition to Eqs. (5), wave equations with right-hand sides of the form suggested by the integrands after taking the operator $\nabla\times\nabla\times$ outside the integral sign:

$$\nabla\times\nabla\times\mathbf{E}-k^2\mathbf{E}=4\pi gk^2\left[\mathbf{P}-\nabla\cdot\hat{Q}+\frac{i}{k}\nabla\times\mathbf{M}\right] , \tag{12a}$$

$$\nabla\times\nabla\times\mathbf{H}-k^2\mathbf{H}=4\pi gk^2\left[\mathbf{M}+\frac{i}{k}\nabla\times\nabla\cdot\hat{Q}-\frac{i}{k}\nabla\times\mathbf{P}\right] , \tag{12b}$$

where g is a freely adjustable parameter. Its value will be chosen in such a way as to satisfy Eqs. (12) and (5) simultaneously.

By using the identities

$$[\nabla\cdot\hat{Q}(\mathbf{r}')G]_j=-Q_{kj}\frac{\partial G}{\partial x_k'}=-\frac{\partial}{\partial x_k'}(Q_{kj}G)+G\frac{\partial}{\partial x_k'}Q_{kj} =[-\nabla'\cdot(\hat{Q}G)+G\nabla'\cdot\hat{Q}]_j , \tag{13a}$$

$$[\nabla\times\mathbf{M}(\mathbf{r}')G]_j=-\epsilon_{jkp}M_p\frac{\partial G}{\partial x_k'} =[-\nabla'\times(\mathbf{M}G)+G\nabla'\times\mathbf{M}]_j , \tag{13b}$$

where $\nabla' G=-\nabla G$, and ∇' indicates the differentiation over $\mathbf{r}'$, the equalities which follow from (13) and the equation $\Delta G+k^2G=0$ for $\mathbf{r}\neq\mathbf{r}'$, we get

$$G(\mathbf{P}+ik\nabla'\times\mathbf{M}-\nabla'\cdot\mathbf{Q})=(\nabla'\times\nabla'\times\mathbf{E})G+\mathbf{E}\Delta' G . \tag{14}$$

Taking into account the identities

$$\begin{aligned}
&G\nabla'\nabla'\cdot\mathbf{E}=\nabla'(G\nabla'\cdot\mathbf{E})-(\nabla' G)(\nabla'\cdot\mathbf{E}) ,\\
&\nabla\int_\sigma^\Sigma G\nabla'\cdot\mathbf{E}d^3\mathbf{r}'=\int_\sigma^\Sigma(\nabla G)(\nabla'\cdot\mathbf{E})d^3\mathbf{r}'+\int_\sigma \mathbf{n}G\nabla'\cdot\mathbf{E}d^2\mathbf{r}_\sigma ,\\
&\int_\sigma^\Sigma\nabla'(G\nabla'\cdot\mathbf{E})d^3\mathbf{r}'=\int_{\Sigma+\sigma}\mathbf{n}G\nabla'\cdot\mathbf{E}d^3\mathbf{r}_\sigma ,\\
&\int_\sigma^\Sigma G\nabla'\nabla'\cdot\mathbf{E}d^3\mathbf{r}'=\int_\Sigma\mathbf{n}_\Sigma G\nabla'\cdot\mathbf{E}d^2\mathbf{r}_\Sigma+\nabla\int_\sigma^\Sigma G\nabla'\cdot\mathbf{E}d^3\mathbf{r}' ,\\
&\nabla\times\int_\sigma^\Sigma G\nabla'\nabla'\cdot\mathbf{E}d^3\mathbf{r}'=\nabla\times\int_\Sigma\mathbf{n}_\Sigma G\nabla'\cdot\mathbf{E}d^2\mathbf{r}'_\Sigma .
\end{aligned} \tag{15}$$

Here Σ is the boundary of the volume of integration, $\mathbf{n}$ is the unit vector normal to the surface Σ or σ, and $\mathbf{n}_\Sigma$ is the unit vector normal to the surface Σ. We used also the well-known relation

$$\int\frac{\partial}{\partial x_i}fd^3\mathbf{r}=\int_\Sigma(\mathbf{n}_\Sigma)_i fd^2\mathbf{r}_\Sigma . \tag{16}$$

As a result all volume integrals are reduced to the Σ or σ surface integrals. The σ-surface integrals are calculated directly. Then by use of Eq. (12) we obtain, after a rather tedious calculation, the equations

$$\left[1-\frac{1}{g}\right]\mathbf{E}=\left[\hat{\beta}+\hat{\gamma}+\frac{4\pi}{3}\right]\cdot\mathbf{P}+\frac{1}{b}(\hat{\eta}+\hat{\xi}):\hat{Q}+ikb(\hat{\beta}_M+\hat{\gamma}_M)\cdot\mathbf{M}$$
$$+b(\hat{\beta}_1+\hat{\gamma}_1):(\nabla\mathbf{P})+\left[\hat{\eta}_1+\hat{\xi}_1-\frac{8\pi}{5}\hat{\delta}\right]:(\nabla\hat{Q})+\mathbf{E}_i$$
$$+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2 g}\left[\mathbf{E}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{E}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E}\right]+G(\hat{Q}\cdot\mathbf{n}_\Sigma+\frac{i}{k}[\mathbf{M}x\mathbf{n}_\Sigma])\right]d^2\mathbf{r}_\Sigma\ , \quad (17a)$$

$$\left[1-\frac{1}{g}\right]\mathbf{H}=\left[\hat{\beta}+\hat{\gamma}+\frac{4\pi}{3}\right]\cdot\mathbf{M}+ik(\hat{\eta}_M+\hat{\xi}_M):\hat{Q}-ikb(\hat{\beta}_M+\hat{\gamma}_M)\cdot\mathbf{P}+b(\hat{\beta}_1+\hat{\gamma}_1):(\nabla\mathbf{M})+\mathbf{H}_i$$
$$+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2 g}\left[\mathbf{H}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{H}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{H}\right]\right.$$
$$\left.-\frac{i}{k}([\mathbf{n}_\Sigma\cdot\hat{Q}\times\nabla'G]+G[\mathbf{n}_\Sigma\times\nabla'\cdot\hat{Q}]+G[\mathbf{P}\times\mathbf{n}_\Sigma])\right]d^2r_\Sigma\ . \quad (17b)$$

Now it is evident that if we choose the values of the free parameters in the following way:

$$g=1,\ \hat{\beta}=-\frac{4\pi}{3}-\hat{\gamma},\ \hat{\beta}_M=-\hat{\gamma}_M,\ \hat{\beta}_1=-\hat{\gamma}_1\ ,$$
$$\hat{\eta}=-\hat{\xi},\ \hat{\eta}_M=-\hat{\xi}_M,\ \hat{\eta}_1=+\frac{8\pi}{5}\hat{\delta}-\hat{\xi}_1\ , \quad (18)$$

where $\hat{\delta}$ is a symmetric unit tensor of fourth rank over two pairs of indices

$$(\hat{\delta})_{ijke}=\delta_{ik}\delta_{je},\ \ \hat{\delta}:(\nabla\hat{Q})=\nabla\cdot\hat{Q}\ , \quad (19)$$

then all the extraintegral terms in (17), except $\mathbf{E}_i$, vanish. As a consequence, Eqs. (17) take the form

$$\mathbf{E}_i+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2}\left[\mathbf{E}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{E}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E}\right]+G\left[\hat{Q}\cdot\mathbf{n}_\Sigma+\frac{i}{k}[\mathbf{M}\times\mathbf{n}_\Sigma]\right]\right]d^2r_\Sigma=0\ , \quad (20a)$$

$$\mathbf{H}_i+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2}\left[\mathbf{H}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{H}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\mathbf{H}\right]-\frac{i}{k}([\mathbf{n}_\Sigma\cdot\hat{Q}\times\nabla'G]\right. \quad (20b)$$
$$\left.+G[\mathbf{n}_\Sigma\times\nabla'\cdot\hat{Q}]+G[\mathbf{P}\times\mathbf{n}_\Sigma])\right]d^2r_\Sigma=0\ .$$

This is the extinction theorem for the anisotropic medium, with the quadrupole and magnetic-dipole mechanisms of nonlinearity.

All these results hold true for the spatially inhomogeneous media as well, but only if the characteristic size of the inhomogeneity is large in comparison with the distance between the oscillators. Therefore, when the medium boundary is "smeared," so that the thickness of the transition layer is large compared with b, the Lorentz's sphere approach may also be applied to the boundary radiators. Thus the boundary Σ can be removed outside of the medium to a region where $\hat{Q}$, $\mathbf{M}$, and $\mathbf{P}$ equal zero. In this case, the extinction theorem takes exactly the well-known form

$$\mathbf{E}_i+\frac{1}{4\pi k^2}\nabla\times\nabla\times\int_\Sigma\left[\mathbf{E}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{E}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E}\right]d^2\mathbf{r}_\Sigma=0\ , \quad (21a)$$

$$\mathbf{H}_i+\frac{1}{4\pi k^2}\nabla\times\nabla\times\int_\Sigma\left[\mathbf{H}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{H}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{H}\right]d^2\mathbf{r}_\Sigma=0\ , \quad (21b)$$

In the limit of a very sharp boundary (compared to b), Eqs. (20) must be corrected, since in this case the contribution of the surface layer, with the thickness less than the Lorentz's sphere radius, must be calculated separately. Obviously, this contribution will depend upon the microscopic relief and surface structure of the medium. In this case the influence of the $\mathbf{P}$, $\mathbf{M}$, and $\hat{Q}$ surface densities may be important.

III. FIELD OUTSIDE A MEDIUM AND MAXWELL EQUATIONS

We next find the fields outside the medium and, in particular, the reflected waves. It is necessary to start from expressions (5) and (10) once more, and reduce volume integrals to surface integrals. With the observation point situated outside the medium, integration takes place over the entire volume. Therefore, the integration and differentiation operations are commutative. As a result, instead of (20) we obtain the formulas

$$\mathbf{E}=\mathbf{E}_i+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2}\left[\mathbf{E}\frac{\partial G}{\partial \nu}-G\frac{\partial \mathbf{E}}{\partial \nu}+G\mathbf{n}_\Sigma(\nabla'\cdot\mathbf{E})\right]+G(\hat{Q}\cdot\mathbf{n}_\Sigma+\frac{i}{k}[\mathbf{M}\times\mathbf{n}_\Sigma])\right]d^2\mathbf{r}_\Sigma\ , \tag{22a}$$

$$\mathbf{H}=\mathbf{H}_i+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2}\left[\mathbf{H}\frac{\partial G}{\partial \nu}-G\frac{\partial \mathbf{H}}{\partial \nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{H}\right]\right.$$
$$\left.-\frac{i}{k}\left[\left[\mathbf{n}_\Sigma\cdot\hat{Q}\times\nabla' G\right]+G\left[\mathbf{n}_\Sigma\times\nabla'\cdot\hat{Q}\right]+G\left[\mathbf{P}\times\mathbf{n}_\Sigma\right]\right]\right]d^2\mathbf{r}_\Sigma\ . \tag{22b}$$

For the medium with the "smeared boundary," Eqs. (22) may be rewritten in the form (21)

$$\mathbf{E}=\mathbf{E}_i+\frac{1}{4\pi k^2}\nabla\times\nabla\times\int_\Sigma\left[\mathbf{E}\frac{\partial G}{\partial \nu}-G\frac{\partial \mathbf{E}}{\partial \nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E}\right]d^2\mathbf{r}_\Sigma\ , \tag{23a}$$

$$\mathbf{H}=\mathbf{H}_i+\frac{1}{4\pi k^2}\nabla\times\nabla\times\int_\Sigma\left[\mathbf{H}\frac{\partial G}{\partial \nu}-G\frac{\partial \mathbf{H}}{\partial \nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{H}\right]d^2\mathbf{r}_\Sigma\ . \tag{23b}$$

By putting Eqs. (18) into formulas (10), we obtain the connection formulas between **E**, **H** and **E**$'$, **H**$'$ fields:

$$\mathbf{E}=\mathbf{E}'-\left[\frac{4\pi}{3}+\hat{\gamma}\right]\cdot\mathbf{P}-\frac{1}{b}\hat{\xi}:\hat{Q}-ikb\hat{\gamma}_M\cdot\mathbf{M}-b\hat{\gamma}_1:(\nabla\mathbf{P})+\frac{8\pi}{5}\nabla\cdot\hat{Q}-\hat{\xi}_1\vdots(\nabla\hat{Q})\ , \tag{24a}$$

$$\mathbf{H}=\mathbf{H}'-\left[\frac{4\pi}{3}+\hat{\gamma}\right]\cdot\mathbf{M}-ik\zeta_M:\hat{Q}+ikb\hat{\gamma}_M\cdot\mathbf{P}-b\hat{\gamma}_1:(\nabla\mathbf{M})\ . \tag{24b}$$

Finally, performing the corresponding substitution of the variables in Eqs. (5),

$$\mathbf{E}=\mathbf{E}_i-\frac{4\pi}{3}\mathbf{P}+\frac{8\pi}{5}\nabla\cdot\hat{Q}+\int_\sigma^\Sigma(\nabla\times\nabla\times\mathbf{P}G-\nabla\times\nabla\times\nabla\cdot\hat{Q}G+ik\nabla\times\mathbf{M}G)d^3\mathbf{r}'\ , \tag{25a}$$

$$\mathbf{H}=\mathbf{H}_i-\frac{4\pi}{3}\mathbf{M}+\int_\sigma^\Sigma(\nabla\times\nabla\times\mathbf{M}G+ik\nabla\times\nabla\cdot\hat{Q}G-ik\nabla\times\mathbf{P}G)d^3\mathbf{r}'\ . \tag{25b}$$

Now by the use of Eqs. (B4) of Appendix B, we can obtain the relationships between **E** and **H**, which, in fact, are the macroscopic Maxwell equations:

$$\begin{aligned}&\nabla\times\mathbf{E}=ik\mathbf{B}=-\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t},\quad \nabla\cdot\mathbf{D}=0\ ,\\&\nabla\times\mathbf{H}=-ik\mathbf{D}=\frac{1}{c}\frac{\partial\mathbf{D}}{\partial t},\quad \nabla\cdot\mathbf{B}=0\ ,\\&\mathbf{D}\equiv\mathbf{E}+4\pi\mathbf{P}-4\pi\nabla\cdot\hat{Q}\ ,\\&\mathbf{B}\equiv\mathbf{H}+4\pi\mathbf{M}\ .\end{aligned} \tag{26}$$

The expression for **D** is identical to result obtained in [13] for the case of the continuous distribution of the charges.

IV. THE LOCAL-FIELD FACTORS FOR AN ANISOTROPIC MEDIUM AND FOR AN ELECTRICALLY AND MAGNETICALLY ISOTROPIC MEDIUM

Finally, we present the macroscopic wave equation in which, for the medium with the linear polarizability $\hat{\alpha}$ and the density of the elementary oscillators N, the polarization term $\mathbf{P}^L=N\hat{\alpha}\mathbf{E}'$ (which is linearly dependent on $\mathbf{E}'$) contributes to the velocity of the propagation. For this it is necessary by means of Eq. (24a) to express $\mathbf{E}'$ through **E** in the equality

$$\mathbf{P}=\mathbf{P}^L+\mathbf{P}^{\mathrm{NL}}=N\hat{\alpha}\mathbf{E}'+\mathbf{P}^{\mathrm{NL}}\ , \tag{27}$$

and to substitute **P** into the wave equation (12a). As a result, we get

$$\nabla\times\nabla\times\mathbf{E}-k^2\hat{\epsilon}\cdot\mathbf{E}=4\pi k^2\left[\hat{f}_p\cdot\mathbf{P}^{\mathrm{NL}}+\hat{f}_Q\cdot(\nabla\hat{Q})+\frac{i}{k}\nabla\times\mathbf{M}-\hat{f}_p\cdot N\hat{\alpha}\cdot\mathbf{F}\right]\ , \tag{28a}$$

$$\hat{\epsilon}=1+4\pi\left[1-\frac{4\pi}{3}N\hat{\alpha}-N\hat{\alpha}\cdot\hat{\gamma}\right]^{-1}\cdot N\hat{\alpha}\ , \tag{28b}$$

$$\hat{f}_p=\left[1-\frac{4\pi}{3}N\hat{\alpha}-N\hat{\alpha}\cdot\hat{\gamma}\right]^{-1}\ , \tag{28c}$$

$$\hat{f}_Q=\hat{f}_p\cdot\left[\left[1-\frac{4\pi}{15}N\hat{\alpha}-N\hat{\alpha}\cdot\hat{\gamma}\right]\cdot\hat{\delta}-N\hat{\alpha}\cdot\hat{\xi}_1\right]\ , \tag{28d}$$

$$F=\frac{1}{b}\hat{\xi}:\hat{Q}+ikb\hat{\gamma}_M\cdot\mathbf{M}+f_p\hat{\gamma}_1:(\nabla\mathbf{P})\ , \tag{28e}$$

where $\hat{\epsilon}$ is the dielectric permittivity tensor.

The separation of the terms in $\hat{Q}$ and **M**, linear in $\mathbf{E}'$ and $\mathbf{H}'$, for the case of an isotropic spatially homogeneous medium leads to not very complex formulas. Taking into account the symmetrical properties of the medium, we come to the following expression for the linear part $\hat{Q}^L$ of the tensor $\hat{Q}$:

$$(\hat{Q}^L)_{ij}=\frac{N\alpha_Q}{k^2}\left[\frac{\partial E'_i}{\partial x_j}+\frac{\partial E'_j}{\partial x_i}\right]. \tag{29}$$

Obviously, for a given oscillator only the field in its vicinity is essential. Therefore, differentiation in (29) is performed over the coordinates of the points near the oscillator, where $\mathbf{E}'$ and $\mathbf{H}'$ satisfy the microscopic Lorentz equations:

$$\nabla\times\mathbf{E}'=ik\mathbf{H}',\quad \nabla\times\mathbf{H}'=-ik\mathbf{E}',\quad \nabla\cdot\mathbf{E}'=\nabla\cdot\mathbf{H}'=0\ . \tag{30}$$

The requirement of the vector equalities covariance in the case of linear dependencies $\mathbf{P}$ on $\mathbf{H}'$ and $\mathbf{M}$ on $\mathbf{E}'$ for an isotropic medium leads to the expression

$$\begin{aligned}\mathbf{P}^L&=N\alpha\mathbf{E}'+\frac{i}{k}N\alpha_H\nabla\times\mathbf{H}'\ ,\\ \mathbf{M}^L&=N\alpha_M\mathbf{H}'+\frac{i}{k}N\alpha_E\nabla\times\mathbf{E}'\ .\end{aligned} \tag{31}$$

Here differentiation is performed in the same sense as in Eqs. (29) and (30).

With the admission of Eq. (30), it is evident that the dependencies of $\mathbf{P}^L$ on $\mathbf{H}'$ and $\mathbf{M}^L$ on $\mathbf{E}'$ only renormalize the constants α and α_M. As a result, for an arbitrary isotropic medium we obtain

$$\begin{aligned}&\mathbf{P}=N\alpha\mathbf{E}'+\mathbf{P}^{\mathrm{NL}},\quad \nabla\cdot\hat{Q}=-N\alpha_Q\mathbf{E}'+\nabla\cdot\hat{Q}^{\mathrm{NL}}\ ,\\ &\mathbf{M}=N\alpha_M\mathbf{H}'+\mathbf{M}^{\mathrm{NL}}\ ,\end{aligned} \tag{32}$$

where the constants α_Q and α_M may be termed quadrupole and magnetic "polarizabilities" of the medium.

Now return to the question about macroscopic properties of media. For an isotropic medium, Eqs. (24) take the form

$$\mathbf{E}=\mathbf{E}'-\frac{4\pi}{3}\mathbf{P}+\frac{8\pi}{5}\nabla\cdot\hat{Q},\quad \mathbf{H}=\mathbf{H}'-\frac{4\pi}{3}\mathbf{M}\ . \tag{33}$$

With substitution of Eq. (32) into (33) and into the wave equations (12), it is necessary to take into account the following circumstance: Unlike the case of Eqs. (29)–(31) on differentiations of the quantities $\mathbf{P}$, $\hat{Q}$, and $\mathbf{M}$ the radius vector $\mathbf{r}'$ now changes from one oscillator coordinate to the next, etc. (but it never lies between the oscillators). As a result a "macroscopic" derivative arises. The quantities $\mathbf{E}'$ and $\mathbf{H}'$ in Eq. (33) and the right-hand sides of Eqs. (12) are differentiated in exactly this way. With the differentiation defined in such a manner, obviously the quantities $\mathbf{E}'$ and $\mathbf{H}'$ are compatible neither with Eqs. (30) nor with Eq. (26). Therefore, for the calculation of the derivatives in Eqs. (33) and (12) it is essential to pass from $\mathbf{E}'$ and $\mathbf{H}'$ to $\mathbf{E}$ and $\mathbf{H}$. After doing so we can use Eqs. (26), into which only the macroscopic derivatives enter.

By use of the equalities (32) and (33), we obtain

$$\mathbf{E}'=\frac{\mathbf{E}+\frac{4\pi}{3}\mathbf{P}^{\mathrm{NL}}-\frac{8\pi}{5}\nabla\cdot\hat{Q}^{\mathrm{NL}}}{1-\frac{4\pi}{3}N\alpha-\frac{8\pi}{5}N\alpha_Q},\quad \mathbf{H}'=\frac{\mathbf{H}+\frac{4\pi}{3}\mathbf{M}^{\mathrm{NL}}}{1-\frac{4\pi}{3}N\alpha_M}\ . \tag{34}$$

Then the wave equation (12a) for $\mathbf{E}$ takes the form

$$\begin{aligned}\nabla\times\nabla\times\mathbf{E}=k^2&\left[\frac{1+\frac{8\pi}{3}N\alpha_M}{1-\frac{4\pi}{3}N\alpha_M}\left[\frac{1+\frac{8\pi}{3}N\alpha+\frac{12\pi}{5}N\alpha_Q}{1-\frac{4\pi}{3}N\alpha-\frac{8\pi}{5}N\alpha_Q}\mathbf{E}+4\pi\frac{1-\frac{4\pi}{15}N\alpha_Q}{1-\frac{4\pi}{3}N\alpha-\frac{8\pi}{5}N\alpha_Q}\mathbf{P}^{\mathrm{NL}}\right.\right.\\ &\left.\left.-4\pi\frac{1+\frac{4\pi}{15}N\alpha}{1-\frac{4\pi}{3}N\alpha-\frac{8\pi}{5}N\alpha_Q}\nabla\cdot\hat{Q}^{\mathrm{NL}}\right]+4\pi\frac{i}{k}\frac{\nabla\times\mathbf{M}^{\mathrm{NL}}}{1-\frac{4\pi}{3}N\alpha_M}\right]\end{aligned} \tag{35}$$

with the precisely analogous form for $\mathbf{H}$.

By means of formally introducing only two new quantities (instead of the microscopic parameters α, α_Q, and α_M),

$$\epsilon=\frac{1+\frac{8\pi}{3}N\alpha+\frac{12\pi}{5}N\alpha_Q}{1-\frac{4\pi}{3}N\alpha-\frac{8\pi}{5}N\alpha_Q}\ , \tag{36a}$$

$$\mu=\frac{1+\frac{8\pi}{3}N\alpha_M}{1-\frac{4\pi}{3}N\alpha_M}\ , \tag{36b}$$

we convert Eqs. (35) into the wave equations of a standard form with the additional electric-quadrupole and magnetic-dipole terms

$$\begin{aligned}&\nabla\times\nabla\times\mathbf{E}-k^2\mu\epsilon\mathbf{E}\\ &\quad=4\pi k^2\left[\mu\frac{\epsilon+2}{3}\mathbf{P}^{\mathrm{NL}}-\mu\frac{2\epsilon+3}{5}\nabla\cdot\hat{Q}^{\mathrm{NL}}\right.\\ &\qquad\left.+\frac{i}{k}\frac{\mu+2}{3}\nabla\times\mathbf{M}^{\mathrm{NL}}\right],\end{aligned}$$

$$\nabla\times\nabla\times\mathbf{H}-k^2\mu\epsilon\mathbf{H}$$
$$=4\pi k^2\left[\epsilon\frac{\mu+2}{3}\mathbf{M}^{NL}+\frac{i}{k}\frac{2\epsilon+3}{5}\nabla\times\nabla\cdot\hat{Q}^{\rm NL}-\frac{i}{k}\frac{\epsilon+2}{3}\nabla\times\mathbf{P}^{\rm NL}\right] . \quad (37)$$

Thus the well-known local-field factor $f_p=(\epsilon+2/3)$ in the case of quadrupole nonlinear source takes the form

$$f_Q=\frac{2\epsilon+3}{5} , \quad (38)$$

and with concurrent allowance of the electric and magnetic phenomena the factors f_p and f_Q are reduced to

$$f_p=\mu\frac{\epsilon+2}{3} \quad \text{and} \quad f_Q=\mu\frac{2\epsilon+3}{5} . \quad (39a)$$

Presumably in a general case of $2n$-multipole radiators local-field factor f_{2n} takes the form

$$f_{2n}=\mu\frac{n\epsilon+n+1}{2n+1} . \quad (39b)$$

V. DISCUSSION

In the special case of the isotropic linear medium, Eqs. (28b) and (36a) immediately reduce to the Lorentz-Lorenz formula

$$\frac{n^2-1}{n^2+2}=\frac{4\pi}{3}N\alpha, \quad n^2=\epsilon . \quad (40)$$

At the same time, due to the vanishing of the third integrand term (since div$\mathbf{E}=0$) the extinction theorem formulation (21) becomes equivalent to the classical EOET statement [1]. The sole distinction lies in the fact that in Eq. (21) the macroscopic field enters, whereas in EOET the linear polarization is proportional to $\mathbf{E}'$ as well as $\mathbf{E}$. With allowance for these respective factors, both the extinction theorem statements become identical.

Proper accounting of the quadrupole radiation in the framework of linear optics for $\mu=1$ gives complete agreement of Eq. (36a) for a macroscopic refractive index with the expression given in Wierzbicki's paper [6] for the case of a plane wave propagating in the semi-infinite, isotropic, homogeneous, linear medium. After substitution of variables (33) and some mathematical transformations, a highly cumbersome expression for an extinction theorem in the paper [6] transforms into the universal formula (20a).

Consider a special case of the nonlinear homogeneous isotropic medium for which the nonlinear part of the polarization $\mathbf{P}^{\rm NL}$ satisfies the wave equation. In distinction to the work of Ref. [7] and following Refs. [11,12], we take here as a wave equation the expression

$$\nabla\times\nabla\times\mathbf{P}^{\rm NL}-\epsilon_s k^2\mathbf{P}^{\rm NL}=0 , \quad (41)$$

but not the form

$$\Delta\mathbf{P}^{\rm NL}-\epsilon_s k^2\mathbf{P}^{\rm NL}=0 . \quad (42)$$

Here $c/\sqrt{\epsilon_s}$ is the velocity of the nonlinear source wave. An approach with the use of Eq. (42) will be discussed later. Let us assume now that now only $\mathbf{E}$ but the field $\mathbf{E}'$ also satisfies the wave equation

$$\nabla\times\nabla\times\mathbf{E}'-\epsilon k^2\mathbf{E}'=4\pi k^2 f_p'\mathbf{P}^{\rm NL} . \quad (43)$$

A volume integral in Eq. (5) may be reduced to a surface integral without any preliminary substitution of the variables. Then Eq. (5) will be satisfied by proper choice of only two (ϵ and f_p') but not three (β, ϵ, f_p or g) parameters, as takes place in a general case:

$$\left[\frac{4\pi}{3}N\alpha\frac{\epsilon+2}{\epsilon-1}-1\right]\mathbf{E}'+\frac{4\pi}{\epsilon_s^2-1}\left[\frac{\epsilon_s^2+2}{3}-\frac{N\alpha}{\epsilon^2-1}f_p'\right]\mathbf{P}^{\rm NL}+\mathbf{E}_i+\frac{1}{k^2}\nabla\times\nabla\times\int_\Sigma\left[\frac{\partial G}{\partial\nu}-G\frac{\partial}{\partial\nu}+\mathbf{n}_\Sigma\nabla'\right]$$
$$\times\left[\frac{N\alpha}{\epsilon-1}\mathbf{E}'+\frac{1}{\epsilon_s-1}\left[1-\frac{4\pi N\alpha f_p'}{\epsilon-1}\right]\mathbf{P}^{\rm NL}\right]d^2\mathbf{r}_\Sigma=0 . \quad (44)$$

As a result we come to the Lorentz-Lorenz formula (40), and the parameter f_p' takes the form

$$f_p'=\frac{\epsilon-1}{4\pi N\alpha}\frac{\epsilon_s+2}{3}=\frac{\epsilon_s+2}{3}\frac{\epsilon+2}{3} . \quad (45)$$

In addition, we obtain the following expression for an extinction theorem:

$$\mathbf{E}_i+\frac{1}{4\pi k^2}\nabla\times\nabla\times\int_\Sigma\left[\frac{\partial G}{\partial\nu}-G\frac{\partial}{\partial\nu}\nabla'\cdot\right]\left[\frac{3}{\epsilon+2}\mathbf{E}'-\frac{4\pi}{3}\mathbf{P}^{\rm NL}\right]d^2\mathbf{r}_\Sigma=0 . \quad (46)$$

By using Eqs. (33) and (40) and neglecting the quadrupolar radiation, Eq. (46) reduces to a universal form (21a). It is easy to verify that appearance of the "extra" factor $(\epsilon_s+2)/3$ is connected with the distinction between $\mathbf{E}$ and $\mathbf{E}'$ in Eqs. (37) and (43). The local factor f_p' turns into f_p on changing from $\mathbf{E}$ to $\mathbf{E}'$ in these equations with use of Eqs. (33) and (34). Bloembergen and Pershan [7], who for the first time considered the case of nonlinear polarization, analyzed model (42), but not (41). Due to this fact they obtained an additional term in Eq. (44) [Eq. (7.5) in Ref. [7]):

$$\frac{4\pi}{(\epsilon_s-1)k^2}\nabla\nabla\cdot(\mathbf{P}^{\rm NL}+N\alpha\mathbf{Q}^a)\ ,$$

where $\mathbf{Q}^a$ and $\mathbf{Q}^b$ are the induced and free parts of the wave produced by the nonlinear polarization and the incident field $\mathbf{E}_i(\mathbf{E}'=\mathbf{Q}^a+\mathbf{Q}^b)$. Due to this fact the cases of the transverse and longitudinal nonlinear polarizations were analyzed in Ref. [7] separately. For the transverse waves models (41) and (42) are identical. As for the case of the longitudinal wave from Eqs. (32), (33), and (37a), when $\hat{Q}=0$ and $\mathbf{M}=0$, we immediately come to the formula

$$\mathbf{Q}^a=-\frac{8\pi}{9}\frac{\epsilon+2}{\epsilon}\mathbf{P}^{\rm NL}=-\frac{2}{3}\frac{\epsilon+2}{3}\frac{4\pi\mathbf{P}^{\rm NL}}{\epsilon} \tag{47}$$

of Ref. [7]. The "extra" [in comparison with $(\epsilon+2)/3$] factor $\frac{2}{3}$ appears here due to the difference between $\mathbf{E}$ and $\mathbf{E}'$ in this case.

As well as in the above-mentioned case, quite an intricate form of the extinction theorem [Eq. (7.6) in Ref. [7]]

$$\mathbf{Q}^{(i)}+\left[\frac{c}{\omega}\right]^2\nabla\times\nabla\times\int_\Sigma ds'\cdot\left\{[\nabla' G(R)]\left[\frac{N\alpha}{\epsilon-1}\mathbf{Q}^b(\mathbf{r}')+\frac{N\alpha\mathbf{Q}^a(\mathbf{r}')+\mathbf{F}(\mathbf{r}')}{\epsilon_s-1}\right]\right.$$
$$\left.-G(R)\nabla'\left[\frac{N\alpha}{\epsilon-1}\mathbf{Q}^b(\mathbf{R}')+\frac{N\alpha\mathbf{Q}^a(\mathbf{r}')+\mathbf{F}(\mathbf{r}')}{\epsilon_s-1}\right]\right\}\ ,$$

reduces to the universal expression (21a) if one takes into account the identity $\mathbf{E}'=\mathbf{Q}^a+\mathbf{Q}^b$ and performs substitutions (33) and (34).

As an illustration, consider a rather elementary problem applying the Lorentz sphere method in the case of a medium with a not discretely but continuously distributed charge density. Then the Lorentz's sphere radius has no lower limit. Obviously, in this case Eqs. (5) hold true. By limiting ourselves for simplicity to the case $\hat{Q}=0$ and $\mathbf{M}=0$ we calculate the field in the center of the Lorentz sphere. This field $\mathbf{E}^{(\sigma)}$ is equivalent to that of two uniformly and oppositely charged balls with the bulk densities ρ, that are displaced by a distance $l \ll a$, as shown in Fig. 1. Evidently,

$$\frac{4\pi}{3}a^3\rho l=\frac{4\pi}{3}a^3P\ . \tag{48}$$

Apparently the product of ρ into l must correspond to a given electric-dipole moment density. For $l<a$ the field in the center of symmetry of such a system is the easiest to calculate. Namely, it is $\mathbf{E}^{(\sigma)}$:

$$\mathbf{E}^{(\sigma)}=-\frac{4\pi}{3}\rho\mathbf{l}=-\frac{4\pi}{3}\mathbf{P}\ . \tag{49}$$

In such a way a medium with a continuous distribution of charges formally is equivalent to a certain "anisotropic" medium with the tensor

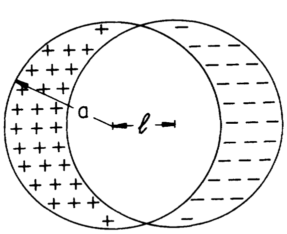

FIG. 1. The contribution of the continuous dipole medium from the interior of Lorentz's sphere is equivalent to the field of two homogeneous and oppositely charged balls. Note that in the whole intersection region of these balls the field is uniform.

$$\hat{\gamma}=-\frac{4\pi}{3}\ , \tag{50}$$

and then from general formulas (18) and (28) it immediately follows that

$$\hat{\beta}=0,\ \ \hat{\mathbf{E}}=\hat{\mathbf{E}}',\ \ \epsilon=1+4\pi N\hat{\alpha}=1+4\pi\hat{\chi},\ \ f_p=1\ , \tag{51}$$

where $\hat{\chi}$ is the linear susceptibility.

Thus we come to a natural result that for a medium with a continuously distributed charge density the formally introduced macroscopic field coincides with the acting one. As regards the extinction theorem, the general expression (20a) holds true in this case as well. For the discrete-oscillator medium no additional bulk or surface currents (the parameters k and j in the notations of Ref. [10]) may exist, other than those described by the variables $\mathbf{P}$, $\hat{Q}$, and $\mathbf{M}$. In this case, and when $\hat{Q}$ equals zero, our results coincide with the results of Refs. [8,9] and with Ref. [10], formula (28).

VI. CONCLUSION

The general idea of substitution of variables into the integral equations (5) of molecular optics, which corresponds to passing from the acting fields $\mathbf{E}'$, $\mathbf{H}'$ to the macroscopic fields $\mathbf{E}$, $\mathbf{H}$, allowed us, under the most general assumptions about nonlinear, anisotropic, spatially inhomogeneous dipole, quadrupolar, and magnetic-dipole media of arbitrary shape, to solve the following problems.

(i) The matching of two approaches of the radiation propagation into the media (Maxwell's equations approach for media with the continuous charge-density distribution and the integral-equation approach for media with a discrete charge-density distribution).

(ii) To formulate an extinction theorem in a general and compact form.

(iii) To obtain universal connection formulas for the characteristic fields and the microscopic and macroscopic quantities of the medium in a general case.

The approach developed gives results coinciding with the previous ones in all special cases. The common property of the previous approximations and assumptions becomes clear: Earlier it was possible to solve similar problems without the substitution of variables (10) only in particular cases when, for one or another concrete reason, the acting field $\mathbf{E}'$ satisfies the Maxwell equations at the same time as the macroscopic field $\mathbf{E}$ satisfies it. The MIE allows us to combine into a single framework both approaches for deduction of the extinction theorem, which correspond to two different models (microscopic approach [1–7,11], and the Maxwell-equation approach [8–10]). Though the quantities $\mathbf{E}$ and $\mathbf{H}$ were introduced due to a formal mathematical requirement, in the end these variables acquired a remarkable additional physical meaning. At least, in the case of an electric-dipole medium these quantities coincide exactly with quantities which arise from different considerations about electromagnetic fields registered by macroscopic devices. As is already known [13,14], [see also formulas (50) and (51)], the physical meaning of the connection between $\mathbf{E}$ and $\mathbf{E}'$ becomes evident if we compare the magnitudes of the fields of the two media with the different microstructures (continuous and discrete charge distribution) at the centers of their respective Lorentz spheres. The difference between results in these two cases is caused by the contribution from the structures, continuous and discrete, inside the Lorentz spheres. It should be emphasized that for the medium with a continuous charge distribution the field in the Lorentz sphere center $\mathbf{E}^{(\sigma)}\neq 0$ [formula (50)], whereas under random or a cubic lattice distribution of discrete dipoles such a field $\mathbf{E}^{(\sigma)}$ always equals zero. In this sense a discrete model does not allow one to obtain the limit of a continuous medium under an unrestricted decrease of the distance between the radiators with simultaneous reduction of the size l, each of them in such a way that the inequality $l \ll b$ holds. As a result, assuming that for a continuous medium $\mathbf{E}=\mathbf{E}'$ [see (51)], for the field $\mathbf{E}'$ acting on a given dipole of the discrete medium, we obtain

$$\mathbf{E}'=\mathbf{E}+\frac{4\pi}{3}\mathbf{P}+\hat{\gamma}\cdot\mathbf{P} .$$

Therefore, in the dipole approximation, neglecting the spatial derivatives of $\mathbf{P}$, we have an expression which coincides with the result of the integral-equation approach [formula (24a)]. The above-mentioned impossibility of passage from a discrete to a continuous medium is a remarkable example of how optical phenomena can be sensitive to the medium structure with arbitrarily small characteristic sizes in comparison with the wavelength. Such sensitivity is realized through local-field effects.

A second example is a distinction in the magnitudes of the constants $\hat{\gamma}$ and $\hat{\zeta}$ for media with different symmetries. As follows from Appendix A, tensors $\hat{\gamma}_1$, $\hat{\gamma}_M$, and $\hat{\zeta}$ equal zero for any inversion-symmetry medium; tensors $\hat{\gamma}$ and $\hat{\zeta}_M$ become zero not only for chaotic media but for a cubic lattice as well. Finally, for a chaotic media tensor $\hat{\zeta}_1=0$. For the cubic lattice this tensor is given in Appendix A. These differences show that quadrupole radiation allows one, by use of the optical methods, to measure finer symmetrical properties of the medium than is possible by means of the dipole radiation only [see formulas (A5) and (28)]. It allows one, for example, to distinguish the cubic lattice from a medium with randomly distributed radiators.

It is interesting to note that for magnetic phenomena, in contrast to electric phenomena, the formal substitution (10) together with a requirement that a new variable must satisfy the wave equation, leads *not* to the field averaged over the interatomic distances (i.e., magnetic induction vector $\mathbf{B}$), but to the quantity $\mathbf{H}$ which enters into the Maxwell equations.

With the simultaneous allowance for electric and magnetic linear polarizabilities it becomes evident that in a wonderful manner the quantities ϵ and μ in formula (36), which appear due to a formal mathematical treatment, have the meaning of the electric and magnetic permittivities. As a result the electric and magnetic characteristics of the medium, which were described by the cumbersome formulas (34) and (35), are separated and reduced into two factors ϵ and μ. These factors give the well-known expression for the refractive index: $n^2=\epsilon\mu$. An even stronger and less evident assertion is true: the local-field factors of the nonlinear electric-dipole, electric-quadrupole, and magnetic-dipole "polarizabilities" in the wave equations include only such combinations of the microscopic polarizabilities α, α_Q, and α_M, which reduce to the quantities ϵ and μ [see Eqs. (35) and (37)]. In the case of a nonmagnetic medium we also have reduction to one parameter ϵ instead of two parameters α and α_Q.

Equations (38) and (39) for these factors permit one, in principle, by measurement of density-dependent nonlinear susceptibilities, to establish the nature of the medium nonlinearities, namely to distinguish anharmonic electric-dipole and electric-quadrupolar microscopic mechanisms by means of macroscopic observations. The timeliness of this consideration follows from present experimental reality. It is now possible to arrange conditions when only electric-quadrupolar and magnetic-dipole nonlinear susceptibilities make contributions to the generation of the second harmonic in a solution of biomolecules [15].

The usual physical interpretation of EOET consists in a statement that an extinction of the incident wave takes place due to the surface layer of the dipoles. But such an interpretation is not fully correct, since, in the expression of the EOET there are two integrals and only one of them corresponds to a radiation from the surface dipole layer. Now, when the role of the quadrupolar radiation became clear, the possibility of the unambiguous interpretation of EOET arises. We notice, in particular, that the first integrand term exactly agrees with the field of the quadrupolar component $Q_{st}=E_s n_{\Sigma t}/4\pi k^2$ (the antisymmetric part of this tensor corresponds to the magnetic moment) distributed over the surface. Therefore, it may be said that the incident field is canceled by the effective radiators with the electric-dipole $(\mathbf{n}_\Sigma\nabla\cdot\mathbf{E}-\partial\mathbf{E}/\partial\nu)d^2r_\Sigma/4\pi k^2$ and electric-quadrupolar $\mathbf{E}\mathbf{n}d^2r_\Sigma/4\pi k^2$ moments which are arranged in a surface layer.

All our considerations are valid for the field outside the medium as well. This means that the reflected wave formation takes place in such a manner as if it had occurred in the surface layer only. But the state of these surface radiators ($\hat{P}$ and $\hat{Q}$) is subject to the action of all the rest of the medium.

The calculations in Appendixes A and B were performed with an accuracy to ka and kb terms, inclusive. Hence all our results are valid to an accuracy of the first order in kb terms. The consideration of the terms $(kb)^2$ with allowance for the spatial dispersion effects will be presented elsewhere.

It should be noted that the main results of this work have rather a simple form, and may be used without detailed acquaintance with the mathematical technique. These are the formulas (28) for the dielectric permittivity $\hat{\epsilon}$ and the local-field factors f_p and f_a for an anisotropic nonmagnetic medium, and the expressions (36), (38), and (39) for ϵ, μ, and local-field factors in the case of the isotropic medium. This also applies to an extinction theorem formulas (20) and (21), and formulas (20) and (23) for the reflected wave for an arbitrary anisotropic nonlinear medium.

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APPENDIX A: THE CALCULATION OF TENSORS $\hat{\gamma}$ and $\hat{\xi}$ FOR CHAOTIC MEDIA AND REGULAR CUBIC LATTICE

General requirements for tensor covariance in center symmetric media demand that

$$\hat{\gamma}_1=\hat{\gamma}_M=\hat{\xi}=0 \; . \tag{A1}$$

Next tensors $\hat{\gamma}$, $\hat{\xi}$, and ζ_M are calculated. The idea of a calculation (see, for instance, Ref. [14]) consists of the fact that for the medium with three orthogonal planes of symmetry (rectangular lattice) all odd-powered terms of the Cartesian components $\mathbf{n}_{jl}$ vanish, and we have

$$(\hat{\gamma})_{st}=b^3\sum_j^{\sigma}\frac{3(\mathbf{n}_{jl})_s^2-1}{R_{jl}^3}\delta_{st} \; , \tag{A2a}$$

$$(\hat{\xi}_1)_{stpq}=-b^3\sum_j^{\sigma}\{15[(n_{jl})_s^2(n_{jl})_p^2\delta_{st}\delta_{pq}+(n_{jl})_s^2(n_{jl})_t^2\delta_{sp}\delta_{tq}+(n_{jl})_s^2(n_{jl})_t^2\delta_{sq}\delta_{tp}](1-\tfrac{2}{3}\delta_{st}\delta_{pq}\delta_{sp})-6(n_{jl})_p^2\delta_{st}\delta_{pq}\}\frac{1}{R_{jl}^3} \; , \tag{A2b}$$

$$(\hat{\xi}_M)_{stp}=3b^3\sum_j\epsilon_{stp}(\mathbf{n}_{jl})_t^2/R_{jl}^3 \; . \tag{A2c}$$

For a cubic lattice additional equalities are valid [14]. In this case we have

$$\sum_j f_1([\mathbf{n}_{jl}]_s^2)f_2(R_{jl})=\sum_j f_1([\mathbf{n}_{jl}]_t^2)f_2(R_{jl}) =\sum_j f_1([\mathbf{n}_{jl}]_p^2)f_2(R_{jl}) \; , \tag{A3}$$

where f_1 and f_2 are arbitrary functions. In particular, from Eq. (A3) it follows that

$$\sum_j(\mathbf{n}_{jl})_s^2 f(R_{jl})=\tfrac{1}{3}\sum_j f(R_{jl}) \; . \tag{A4}$$

From (A4), which is valid for a cubic lattice, it follows at once that $\hat{\gamma}=0$, and $\hat{\xi}_M$ is proportional to $\hat{\epsilon}$. Then the contraction $\hat{\xi}_M$ with the symmetric tensor $\hat{Q}$ equals zero. It is equivalent to setting tensor $\hat{\xi}_M=0$ and writing an expression for $\hat{\xi}_1$ in the following form [16]:

$$(\hat{\xi}_1)_{ssss}=-\tfrac{3}{2}(\hat{\xi}_1)_{stts}\Big|_{t\neq s} =\tfrac{9}{2}b^3\sum_j^{\sigma}\frac{1-5(\mathbf{n}_{jl})_s^4}{R_{jl}^3}=-14.01 \; , \tag{A5}$$

whereas the remaining components of the tensor $\hat{\xi}_1$ equal zero. Note that Eq. (A5) converges rapidly approaching the limit value after summation over three or four oscillator shells.

Finally, for a chaotic distribution of the oscillators, and taking advantage of the averaging over an ensemble of the random spatial configurations, summation may be replaced by integration. From symmetry considerations it follows that there will be a zero average for all uniform tensor components. In addition, due to angular dependence $(\mathbf{n}_{jl})_s$, the right-hand side of Eq. (A5) goes to zero after angular integration. As a result, Eqs. (9) follow.

APPENDIX B: FACTORING OF THE OPERATOR $\nabla\times\nabla\times$ OUTSIDE THE INTEGRAL SIGN

We start from an equally [1]

$$\frac{\partial}{\partial x_i}\int_{\sigma}^{\Sigma}Fd^3\mathbf{r}'=\int_{\sigma}^{\Sigma}\frac{\partial F}{\partial x_i}d^3\mathbf{r}'+\int_{\sigma}Fn_i d^2\mathbf{r}' \; , \tag{B1}$$

where $F=F(\mathbf{r},\mathbf{r}')$ is an arbitrary function, and $\mathbf{n}$ is the unit vector normal to the surface σ.

Set $F=f(\mathbf{r}')e^{ikR}/R$ and, taking into account the small size of a, expand $f(\mathbf{r}')$ in a power series in the vicinity of the point $\mathbf{r}$:

$$f(\mathbf{r}')=f(\mathbf{r})-\frac{\partial f}{\partial x_i}n_i a+\frac{\partial^2 f}{2\partial x_i\partial x_j}n_i n_j a^2+\cdots =f(\mathbf{r})-a(\mathbf{n}\cdot\nabla)f+\frac{a^2}{2}(\mathbf{nn}:\nabla\nabla)f+\cdots \; . \tag{B2}$$

Calculating directly integrals over σ in the right-hand of equality (B1), we come to the equations:

$$\int_\sigma fGn_i d^2\mathbf{r}' = fae^{ika}\int n_i d\Omega - \frac{\partial f}{\partial x_j}a^2 e^{ika}\int n_i n_j d\Omega + \frac{1}{2}\frac{\partial^2 f}{\partial x_j \partial x_k}a^3 e^{ika}\int n_i n_j n_k d\Omega + O\left[a^4 \frac{\partial^4 f}{\partial x^4}\right]$$

$$= -\frac{4\pi}{3}a^2 e^{ika}\frac{\partial f}{\partial x_i} + O\left[a^4 \frac{\partial^2 f^4}{\partial x^4}\right] = 0 + O(a^2) , \tag{B3a}$$

$$\int_\sigma f\frac{\partial G}{\partial x_j}n_i d^2\mathbf{r}' = -(1-ika)e^{ika}\left[f\int n_i n_j d\Omega + \frac{a^2}{2}\frac{\partial^2 f}{\partial x_k \partial x_l}\int n_i n_j n_k n_l d\Omega\right] + O\left[a^4\frac{\partial f^4}{\partial x^4}\right] = -\frac{4\pi}{3}f\delta_{ij} , \tag{B3b}$$

$$\int_\sigma \frac{\partial^2 G}{\partial x_j \partial x_k}n_i d^2\mathbf{r}' = \frac{4\pi}{15}\left[2\frac{\partial f}{\partial x_i}\delta_{jk} - 3\left[\frac{\partial f}{\partial x_j}\delta_{ij} + \frac{\partial f}{\partial x_k}\delta_{ij}\right]\right] + O(a^2) , \tag{B3c}$$

$$\int_\sigma f\frac{\partial^3 G}{\partial x_j \partial x_k \partial x_l}n_i d^2\mathbf{r}' = \frac{4\pi}{5}\left[\left[\frac{k^2}{3}f + \frac{1}{7}\frac{\partial^2 f}{\partial x_m^2}\right](\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{2}{7}\left[\frac{\partial^2 f}{\partial x_i \partial x_j}\delta_{kl} + \frac{\partial^2 f}{\partial x_i \partial x_k}\delta_{jl} + \frac{\partial^2 f}{\partial x_i \partial x_l}\delta_{jk}\right]\right.$$

$$\left. - \frac{5}{7}\left[\frac{\partial^2 f}{\partial x_k \partial x_l}\delta_{ij} + \frac{\partial^2 f}{\partial x_j \partial x_l}\delta_{ik} + \frac{\partial^2 f}{\partial x_j \partial x_k}\delta_{il}\right]\right] . \tag{B3d}$$

Formulas (B3) are accurate up to first-order terms in ka, inclusive. Here, just as in Eqs. (6) and (8), appears the factor $(1-ika)e^{ika}$, which equals unity to the same order. By use of Eqs. (B1) and (B2), we obtain equalities which are accurate up to the first-order terms in ka, inclusive:

$$\nabla\times\int_\sigma^\Sigma \mathbf{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\mathbf{f}Gd^3\mathbf{r}' , \tag{B4a}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\times\mathbf{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\mathbf{f}Gd^3\mathbf{r}' + \frac{8\pi}{3}\mathbf{f} , \tag{B4b}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\times\nabla\times\mathbf{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\nabla\times\mathbf{f}Gd^3\mathbf{r}' + \frac{4\pi}{3}\nabla\times\mathbf{f} , \tag{B4c}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\cdot\hat{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' - \frac{4\pi}{3}\hat{\epsilon}:\hat{f} , \tag{B4d}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' + \frac{12\pi}{5}[\nabla\cdot\hat{f} - \tfrac{1}{3}\nabla\mathrm{Tr}\hat{f} + \tfrac{2}{9}\nabla\cdot(\hat{f}^* - \hat{f})] ,$$

$$\nabla\times\int_\sigma^\Sigma \nabla\times\nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' + \frac{8\pi}{5}[\nabla\times\nabla\cdot\hat{f} + \tfrac{1}{2}\nabla\nabla\cdot(\hat{\epsilon}:\hat{f}) - \Delta(\hat{\epsilon}:\hat{f})] . \tag{B4e}$$

With an additional use of Eqs. (B4), we arrive at the desired relationships:

$$\nabla\times\nabla\times\int_\sigma^\Sigma \mathbf{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\mathbf{f}Gd^3\mathbf{r}' + \frac{8\pi}{3}\mathbf{f} , \tag{B5a}$$

$$\nabla\times\nabla\times\int_\sigma^\Sigma \nabla\times\mathbf{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\nabla\times\mathbf{f}Gd^3\mathbf{r}' + 4\pi\nabla\times\mathbf{f} , \tag{B5b}$$

$$\nabla\times\nabla\times\int_\sigma^\Sigma \nabla\cdot\hat{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' + \frac{12\pi}{5}[\nabla\cdot\hat{f} - \tfrac{1}{3}\nabla\,\mathrm{Tr}\hat{f} + \tfrac{1}{3}\nabla\cdot(\hat{f} - \hat{f}^*)] , \tag{B5c}$$

$$\nabla\times\nabla\times\int_\sigma^\Sigma \nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' = \int_\sigma^\Sigma \nabla\times\nabla\times\nabla\times\nabla\cdot\hat{f}Gd^3\mathbf{r}' + 4\pi\nabla\times\nabla\cdot\hat{f} + \frac{4\pi}{3}[\nabla\times\nabla\cdot(\hat{f}^* - \hat{f}) - k^2\hat{\epsilon}:\hat{f}] , \tag{B5d}$$

where $\hat{f}^*$ is for the transposed tensor $\hat{f}$.

In conclusion we see that the effect of factoring the ∇ operators is the appearance of new terms in addition to the integral. All these terms contain factors $(1-ika)e^{ika}$, and all the equations are valid not only in a static-field approximation, but with the additional terms ka, as well.

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