Discreteness and local fields in weakly rarefied media

A. V. Ghiner[1,*] and G. I. Surdutovich[2,†]
[1]Departamento de Fisica, Universidade Federal do Ceara, Caixa Postal 6030, CEP 60450-970 Fortaleza, Ceara, Brazil
and Departamento de Fisica, Universidade Federal do Maranhao, CEP 65085-580 Sao Luis, Maranhao, Brazil
[2]DSIF, FEEC, Caixa Postal 6101, Unicamp, CEP 13083-970 Campinas, Sao Paulo, Brazil
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A generalized method of integral equations (GMIE) is applied to a weakly rarefied (not so dense) medium when the distance b between the neighboring elementary radiators is already not negligibly small in comparison with the wavelength of light λ. In this paper discreteness is treated not as a conceptual idea only but as a quantitatively measured parameter b/λ. It was found that the extinction theorem and Maxwell's equations remain valid even in such conditions. A striking contradiction with the energy conservation law arising with allowance of the radiation damping effect into the Lorentz-Lorenz formula is resolved by means of a proper account of the discreteness of the medium. The approach developed enabled us to calculate the local field factors and dielectric permittivity of a rarefied medium. An essential quantitative and qualitative distinction between the gaslike, jellylike, and a cubic lattice media, customarily treated as optically isotropic, was revealed. For a cubic lattice crystal an optical anisotropy is predicted. The possibility of application of the GMIE to calculation of the integral light scattering in an irregular medium is discussed. Our results may be applied to calculation of the optical properties of some specific types of media, such as a cooled atomic gas, composite materials, and quantum dots structures. [S1063-651X(97)10511-6]

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I. INTRODUCTION

What can we learn about the internal microstructure of an optical medium from macroscopic optical measurements? Microscopic symmetry of ordered media (crystals) manifests itself in an anisotropy of the refractive index. Whereas the internal structure of radiators is taken into account by multipolar expansion, neither radiator size nor lattice grain size ever enters into formulas for the refractive index n of a medium. In optics such an approach is usually well-grounded because of the smallness of these sizes compared with the wavelength λ. Thus, according to the classical Lorentz-Lorenz (LL) formula [1,2], the optical properties of an isotropic medium depend merely on the product of a density N of the radiators and the polarizability α of an isolated radiator. This paper is the third in a series of papers devoted to the study of the the connection of the macroscopic optical properties of the medium with its microstructure, i.e., geometric disposition of the elementary radiators and the distances b between them. In the previous two papers we developed the fundamentals of the theory and applied method of integral equations (MIE) to an arbitrary nonlinear and anisotropic dense medium (paper I [3]) and some two-dimensional and quasi-two-dimensional problems (paper II [4]). In those cases only the unitless geometrical tensor $\hat{\gamma}$ of a lattice and the radiator's polarizability tensor $\hat{\alpha}$, but not a lattice grain size b, determine the local field factor $\hat{f}_p$ and enter into the anisotropic analogues of the LL formula I, II, [5]. The present paper is devoted to a detailed study of the influence of a medium's discreteness (the parameter b) on the optical properties in the case of a weakly rarefied medium.

As is evident from its derivation, the LL formula holds true only for a dense, $N\lambda^3 \gg 1$, medium. Although just a discreteness gives rise to the local field effects ''incorporated'' into the LL formula and anisotropic analogues, these formulas reveal only *the presence* of a medium's discreteness, but not its ''magnitude'' b. Over the last years, an interest has emerged in ''essentially discrete'' media (cooled atomic gas [6], photonic band gap [7] and quantum dots [8] structures), when the distances b between the radiators are not negligible (or small) compared with the wavelength λ. Moreover, a problem arises already in the course of a self-consistent description of an ordinary dense media. This paper pursues a line of thought by Planck and Mandelstam, who first considered a problem of the proper account of the radiation damping (RD) effects in optically homogeneous discrete media [9] (for posterior considerations see Refs. [10, 11]). Really, in accordance with the general concept and concrete derivation of the LL formula for a dense medium, it seems reasonable to substitute into this formula total complex polarizability $\alpha = \alpha' + i\alpha''$ of an isolated elementary radiator with allowance for the contribution of the RD into α'', as was made by Hippel [12]. However, such a natural procedure of an account of the RD effects leads to a paradoxical conclusion that the existence of the nonabsorbing dielectric crystals in frameworks of the classical molecular optics turns out to be impossible [13]. It is worth mentioning that the Clausius-Mossotti formula, the electrostatic counterpart of the LL relation, does not pose such a problem [14], since in electrostatics there are no RD effects. As will be shown in more detail in the course of this paper, the correct consider-

*Permanent address: Automation and Electrometry Institute, 630090 Novosibirsk, Russia.
Electronic address: ghiner@gate.fapema.br
†Corresponding author. Permanent address: Semiconductor Physics Institute, 630090 Novosibirsk, Russia. Electronic address: gregory@dsif.fee.unicamp.br

ation of the medium's discreteness solves the problem. It occurs that for self-consistency of the molecular optics it is indeed necessary to take into account the RD effects. However, such an account of these effects by no means implies a naive *direct* substitution of the imaginary part of α (or tensor polarizability $\hat{\alpha}$) of an isolated oscillator into the LL formula (or its analogs for an anisotropic medium, see I and II). The point is that the RD appears due to retardation in the process of emission of an isolated radiator, so that proper account of *all retardation effects* implies that their allowance be made in the interparticle interactions effects *as well*. In other words, it means breaking with the generally accepted electrostatic approximation in the derivation of the LL formula.

The established way of dealing with the LL relation is to proceed from the macroscopic dielectric function of the equivalent homogeneous medium to the microscopic parameters of this medium. It is the essence of the numerous techniques developed for description of discrete media after the pioneering work by Purcell and Pennypacker, where a continium medium was replaced by an array of point entities (dipoles) [15]. Mathematical sophistry aside, these approaches as well as the equivalent integral forms of Maxwell's equations allow us to more or less exactly restore the point-polarizable entities (atomic polarizabilities) from macroscopic considerations.

A MIE [1] allows us to reverse the order of the conventional approach, i.e., to deduce the macroscopic quantities of the medium from their microscopic counterparts. The standard MIE is the embodiment of the discrete concept and affords one an opportunity for self-consistent description of all optical phenomena in the dense media. By virtue of the medium's ''compactness'' one yields two beneficial advantages: (i) the possibility to consider all the radiators inside the Lorentz sphere (LS), with a certain radius a, as being identical, so that polarization inside the LS may be kept constant; (ii) the possibility to replace the summation over out-of-LS radiators by an integration and thus to produce an integral equation. In this case compactness of the medium is the essential factor since it allows us to impose the macroscopic condition of the greatness of the LS radius compared with the distance b between the radiators (for a crystal b is the cube root of the volume of an elementary cell)

$$a \gg b \tag{1a}$$

and, at the same time, the microscopic homogeneity condition

$$a \ll \lambda . \tag{1b}$$

A rarefied medium poses a nontrivial problem since both of these benefits vanish and the standard MIE cannot be applied. Hence what is required is a judicious generalization of the MIE approach. Here we will develop such a theory. To overcome the first obstacle—nonidentity of the radiators inside the LS—we took into account the variation of the polarization inside the LS:

$$\vec{P}(\vec{r}_j) = \vec{P}(\vec{r}_l) + (\vec{R}_{jl} \cdot \nabla)\vec{P}_{\vec{r}=\vec{r}_e} + \frac{1}{2}(\vec{R}_{jl}\vec{R}_{jl} : \nabla\nabla)\vec{P}_{\vec{r}=\vec{r}_e} \cdots , \tag{2}$$

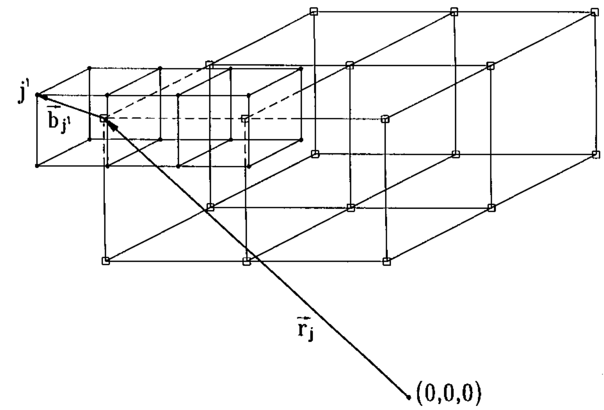

FIG. 1. A splitting procedure for a cubic lattice. □ denotes the position of the primary atoms of the initial lattice and • stands for the secondary atoms of the splitted lattice. Vector $\vec{r}_j$ stands for the jth atom position, vector $\vec{b}_{j'}$ stands for the position of the j'th secondary atom (j' changes from 1 to 8).

where $\vec{R}_{jl} \equiv \vec{r}_j - \vec{r}_l$, R_{jl} is the distance of jth radiator from the LS center, which stands as $\vec{r}_l$. Furthermore, we will account for all such additional terms. The second problem is connected with out-of-LS radiators. For a rarefied medium *one cannot* simply turn the summation into an integration, as is assuming in the case of a dense medium. Really, owing to a finite value of the parameter kb ($k = 2\pi/\lambda$ is the wave number of the light) the contribution to the field in the center of the LS due to of the phase difference between two dipoles situated outside the LS *remains substantional at any distances of these dipoles from the center*. In particular, two dipoles at points j and j', with a finite distance b between them, produce at any observation point ($\vec{r}_l$, for example) a different field as compared with an isolated dipole with the same total dipole moment. To consider these dipoles as if stuck together it is necessary to satisfy not only the condition of their remoteness from the LS center

$$b \ll R_{jl} , \tag{3a}$$

but also the inequality

$$kb \ll 1,$$

i.e,

$$b \ll \lambda . \tag{3b}$$

Therefore, if one intends to account for the discreteness of a medium, one cannot simply pass from the sum to the integral. At first sight this seems like an insuperable obstacle on route to integral equations. However, it is possible (and sufficient) to find the *difference* between such a sum and the integral and so to turn the summation into an integration. For this purpose we have developed a special procedure of ''radiator splitting.'' Its essence is the following. We split each of the elementary radiators into eight smaller ones to obtain a lattice with the period $b/2$ (see Fig. 1). After that we expand the field from the ''split dipoles'' at the observation point by the parameter b/R_{jl} and calculate the difference between local fields $\vec{E}'(\vec{r}_l)$ from the lattices with periods b

and $b/2$ and the same volume densities of the dipole moment (polarization) $\vec{P}$. By iteration of this procedure, the difference of the acting fields from an initial lattice with grain size b and a similar lattice with an arbitrary small b is calculated. It solves the problem of the passage to an integral equation. This is one of the most important end results of the developed theory.

Section II deals with the earlier developed GMIE modified for the rarefied media. In Sec. III the role of the RD effects is considered, whereas in Sec. IV we calculate the optical properties of the regular and random media. For simplicity we limit ourselves, for the most part, to the case of an electric-dipole medium. The calculations for the electric quadrupole and magnetic-dipole media, although much more cumbersome, can be done in a similar way. Such results are present in Sec. V. Section VI states that the extinction theorem and the Maxwell equations remain valid for a weakly rarefied medium. Section VII for a summary and interpretation of the results. Several Appendixes contribute technical details necessary for the clarity of the presentation.

II. GMIE APPROACH MODIFIED FOR A RAREFIED MEDIUM

Let us consider the simplest case of a medium constituted from identical particles in the equivalent conditions. Such a medium has a center of symmetry. Later this fact will be taken into account under expansion of lattice sums. The consideration of more complicated noncentrosymmetric media will be given elsewhere. Later on all the calculations will be performed to third order in the kb parameter since these $(kb)^3$ terms describe the RD effect.

The total account of the discreteness is included in the fundamental equations of the molecular optics for electric $\vec{E}'(\vec{r}_l)$ and magnetic $\vec{H}'(\vec{r}_l)$ local fields acting on a radiator at the point $\vec{r}_l$ with the dipole moment $\vec{p}(\vec{r}_l)$ [1]:

$$\vec{E}'(\vec{r}_l)=\vec{E}_i(\vec{r}_l)+\sum_{j\neq l}\nabla\times\nabla\times\vec{p}(\vec{r}_j)\,\frac{e^{ik|\vec{r}_j-\vec{r}_l|}}{|\vec{r}_j-\vec{r}_l|},\qquad(4a)$$

$$\vec{H}'(\vec{r}_l)=\vec{H}_i(\vec{r}_l)-ik\sum_{j\neq l}\nabla\times\vec{p}(\vec{r}_j)\,\frac{e^{ik|\vec{r}_j-\vec{r}_e|}}{|\vec{r}_j-\vec{r}_l|}.\qquad(4b)$$

Here $\vec{E}_i(\vec{r}_l)$ and $\vec{H}_i(\vec{r}_l)$ are the strengths of the electric and magnetic fields of an incident wave at the point $\vec{r}_l$, j, and l denote the indices of the dipoles.

By use of the splitting procedure (see Appendix A) we pass from the summation in Eq. (4) to the integration outside of the LS and come to the integral equations

$$\vec{E}'(\vec{r})=\vec{E}_i(\vec{r})+\vec{E}_\sigma(\vec{r})+\vec{E}_b(\vec{r})+\int_\sigma^\Sigma\nabla\times\nabla\times\vec{P}G\,d^3\vec{r}\,',\qquad(5a)$$

$$\vec{H}'(\vec{r})=\vec{H}_i(\vec{r})+\vec{H}_\sigma(\vec{r})+\vec{H}_b(\vec{r})-\int_\sigma^\Sigma ik\nabla\times\vec{P}G\,d^3\vec{r}\,',\qquad(5b)$$

where $\vec{P}(\vec{r}\,')=N\vec{p}$ is the polarization of the medium $\vec{E}_\sigma$ and $\vec{H}_\sigma$ are the contributions from the dipoles inside the LS, $G(R)=e^{ikR}/R$ is the Green function of a scalar wave equation $\vec{R}\equiv\vec{r}-\vec{r}\,'$, and Σ is the boundary of the medium. Equations (5) differ from the earlier deduced integral equations (see I, II) by the additional terms $\vec{E}_b$ and $\vec{H}_b$, arising due to the procedure of splitting and accounting a finite value of the parameter kb (see Appendix A).

The splitting procedure produces in Eqs. (5) some additional surface integrals with respect to the polarization $\vec{P}$ and its spatial derivativies. For simplicity, we limit ourselves to the simple case of a medium with the ''blurred boundary'' (see discussion in the paper I and Sec. V of the present paper) when these integral terms go to zero. The general case is considered in Sec. V.

First, we write the expressions for the contributions $\vec{E}_\sigma$ and $\vec{H}_\sigma$ with allowance of the kb terms:

$$\vec{E}_\sigma=\hat{\gamma}_0\cdot\vec{P}+b^2\hat{\gamma}_2\,\vdots\,(\nabla\nabla\vec{P}),\qquad(6a)$$

$$\vec{H}_\sigma=ikb^2\hat{\gamma}_{M1}:(\nabla\vec{P}),\qquad(6b)$$

where, $\hat{\gamma}_0$, $\hat{\gamma}_{M1}$, and $\hat{\gamma}_2$ are the dimensionless second, third, and forth rank tensors, respectively (see Appendix B). These tensors are determined by the spatial distribution of the radiators (for example, the geometry of a crystalline lattice). For a noncentersymmetric medium Eqs. (6a) and (6b) must be complemented by the terms $\hat{\gamma}_1:(\nabla\vec{P})$ and $ikb\hat{\gamma}_{M0}\cdot\vec{P}$ (see I). For a random medium and a cubic lattice the tensors are calculated in Appendix B. For our paramount terms $\vec{E}_b$ and $\vec{H}_b$ we have similar expressions (Appendix A):

$$\vec{E}_b=(kb)^2\hat{\gamma}_{b0}\cdot\vec{P}+b^2\hat{\gamma}_{b2}\,\vdots\,(\nabla\nabla\vec{P}),\qquad(7a)$$

$$\vec{H}_b=ikb^2\hat{\gamma}_{Mb1}:(\nabla\vec{P}).\qquad(7b)$$

Further, we will proceed on the lines of the generalized method of integral equations (GMIE) developed in I and II. Namely, let us rewrite Eqs. (5) in terms of new variables $\vec{E}$ and $\vec{H}$ [compare I, Eqs. (10), and II, Eqs. (13)]:

$$\vec{E}=\vec{E}'+\hat{\beta}_0\cdot\vec{P}+b^2\hat{\beta}_2:(\nabla\nabla\vec{P}),\qquad(8a)$$

$$\vec{H}=\vec{H}'+ikb^2\hat{\beta}_{M1}:(\nabla\vec{P}).\qquad(8b)$$

At this stage tensors $\hat{\beta}_0$, $\hat{\beta}_2$, and $\hat{\beta}_{M1}$ are still free parameters. As was shown earlier in I and II, under a certain choice of the values of these tensors and those variables formally introduced in a remarkable manner acquire the *physical sense of the macroscopic fields* $\vec{E}$ *and* $\vec{H}$. However, in contrast to I and II, there are additional terms $(kb)^2$ which now play an important role. The third order terms, $b^3\nabla\nabla\nabla\vec{P}$ for the electric field and $ikb^3\nabla\nabla\vec{P}$ for the magnetic field, alongside the first order ones $b\nabla\vec{P}$ and $ikb\vec{P}$, vanish due to the symmetric properties of a medium. Taking into account I, Eqs. (11), we may factor the operator $\nabla\times\nabla\times$ outside the integral sign while keeping an accuracy up to the $(kb)^3$ terms (see Appendix C). Suppose now that the variables $\vec{E}$

and $\vec{H}$ satisfy not only Eqs. (5) but also the specific wave equations with nonzero right-hand sides:

$$\nabla\times\nabla\times\vec{E}-k^2\vec{E}=4\pi k^2\vec{P}, \tag{9a}$$

$$\nabla\times\nabla\times\vec{H}-k^2\vec{H}=-4\pi ik\nabla\times\vec{P}, \tag{9b}$$

Using the same method as in I and II (see also Appendix D) we transform volume integrals in Eqs. (5) into the surface ones and thus arrive to the equations

$$\left[\hat{\beta}_0+\frac{4\pi}{3}+\hat{\gamma}_0+(kb)^2\hat{\gamma}_{b0}-\hat{\gamma}_0'\right]\cdot\vec{P}+b^2(\hat{\beta}_2+\hat{\gamma}_2+\hat{\gamma}_{b2}-\hat{\gamma}_2')\,\vdots\,(\nabla\nabla\vec{P})+\vec{E}_i+\frac{\nabla\times\nabla\times}{4\pi k^2}\times\int_\Sigma\left(\vec{E}\,\frac{\partial G}{\partial\nu}-G\,\frac{\partial\vec{E}}{\partial\nu}+G\vec{n}_\Sigma\nabla'\cdot\vec{E}\right)d^2\vec{r}_\Sigma=\vec{0}, \tag{10a}$$

$$ikb^2(\hat{\beta}_{M1}+\hat{\gamma}_{M1}+\hat{\gamma}_{bM1}-\hat{\gamma}_{M1}'):(\nabla\vec{P})+\vec{H}_i+\frac{\nabla\times\nabla\times}{4\pi k^2}\int_\Sigma\left(\vec{H}\,\frac{\partial G}{\partial\nu}-G\,\frac{\partial\vec{H}}{\partial\nu}+G\vec{n}_\Sigma\nabla'\cdot\vec{H}\right)d^2\vec{r}_\Sigma=\vec{0}, \tag{10b}$$

where $\nabla'\equiv\partial/\partial\vec{r}\,'$ and $\vec{n}_\Sigma$ is the unit vector of the outward normal to the boundary Σ,

$$(\hat{\gamma}_0')_{st}=\gamma_0'\delta_{st}, \tag{11a}$$

$$\gamma_0'=\frac{4\pi}{3}(ka)^2-\frac{8\pi}{9}\,i(ka)^3, \tag{11b}$$

$$(\hat{\gamma}_2')_{stpq}=\frac{2\pi}{5}(ka)^2\left(\delta_{st}\delta_{pq}-\frac{1}{3}\,\delta_{sq}\delta_{ip}\right), \tag{11c}$$

$$\hat{\gamma}_2'\,\vdots\,\nabla\nabla\vec{P}=\frac{2\pi}{5}(ka)^2\left(\nabla\nabla\cdot\vec{P}-\frac{1}{3}\,\Delta\vec{P}\right), \tag{11d}$$

$$(\hat{\gamma}_{M1}')_{stp}=-\frac{2\pi}{3}(ka)^2\epsilon_{stp}, \tag{11e}$$

$$\hat{\gamma}_{M1}':\nabla\vec{P}=-\frac{2\pi}{3}(ka)^2\nabla\times\vec{P}. \tag{11f}$$

Here s,t,p,q denote the Cartesian component numbers and ϵ_{stp} is the antisymmetric unit tensor of the third rank. Tensors $\hat{\gamma}_0'$, $\hat{\gamma}_2'$, $\hat{\gamma}_{Ml}'$ arise from the integrals in Eqs. (5) due to the factoring of the operator $\nabla\times\nabla\times$ and the reducing of the integrals to surface ones (see Appendixes C and D).

Now let us choose the values of free parameters $\hat{\beta}_0$, $\hat{\beta}_2$, and $\hat{\beta}_{M1}$ to satisfy Eqs. (9) and (10):

$$\hat{\beta}_0=-\frac{4\pi}{3}-\hat{\gamma}_0^{(0)}-(kb)^2\hat{\gamma}_0^{(2)}+\frac{8\pi}{15}(kb)^2(\hat{\Phi}-2\,\mathrm{Tr}\,\hat{\Phi})+\frac{2}{3}\,i(kb)^3, \tag{12a}$$

$$(\hat{\beta}_2)_{stpq}=-(\hat{\gamma}_2^{(0)})_{stpq}-\frac{8\pi}{105}[6(\hat{\Phi})_{st}\delta_{pq}+3(\hat{\Phi})_{sq}\delta_{tp}-4(\hat{\Phi})_{tp}\delta_{sq}+6(\hat{\Phi})_{pq}\delta_{st}]+\frac{4\pi}{35}(5\delta_{st}\delta_{pq}-\delta_{sq}\delta_{tp})\mathrm{Tr}\,\hat{\Phi}. \tag{12b}$$

$$(\hat{\beta}_{M1})_{stp}=-(\gamma_{M1}^{(0)})_{stp}+\frac{4\pi}{5}\left[\frac{4}{3}\,\hat{\varepsilon}_{stq}(\hat{\Phi})_{qp}-\varepsilon_{stp}\,\mathrm{Tr}\,\hat{\Phi}\right], \tag{12c}$$

$$(\hat{\gamma}_0^{(0)})_{st}=\lim_{A\to\infty}\sum_{j\neq l}^{K_{jl}\leqslant A}\frac{3(\vec{n}_{jl})_s(\vec{n}_{jl})_t-\delta_{st}}{K_{jl}^3}, \tag{12d}$$

$$(\hat{\gamma}_0^{(2)})_{st}=\lim_{A\to\infty}\left[\frac{1}{2}\sum_{j\neq l}^{K_{jl}\leqslant A}\frac{(\vec{n}n_{jl})_s(\vec{n}_{jl})_t+\delta_{st}}{K_{jl}}-\frac{4\pi}{3}\,A^2\delta_{st}\right], \tag{12e}$$

$$(\hat{\gamma}_2^{(0)})_{stpq}=\lim_{A\to\infty}\left[\frac{1}{2}\sum_{j\neq l}^{K_{jl}\leqslant A}\frac{3(\vec{n}_{jl})_s(\vec{n}_{jl})_t(\vec{n}_{jl})_p(\vec{n}_{jl})_q-(\vec{n}_{jl})_t(\vec{n}_{jl})_p\delta_{sq}}{K_{jl}}-\frac{2\pi}{5}\,A^2\left(\delta_{st}\delta_{pq}-\frac{1}{3}\,\delta_{sq}\delta_{tp}\right)\right], \tag{12f}$$

$$(\hat{\gamma}_{M1})_{stp}=\lim_{A\to\infty}\left[\frac{2\pi}{3}\,A^2\varepsilon_{stp}-\varepsilon_{sqp}\sum_{j\neq l}^{K_{jl}\leqslant A}\frac{(\vec{n}_{jl})_q(\vec{n}_{jl})_t}{K_{jl}}\right], \tag{12g}$$

where $K_{jl}\equiv R_{jl}/b$, $A\equiv a/b$, and $\hat{\Phi}$ is the unitless second-rank tensor that arose due to the splitting procedure [see Appendix A, Eqs. (A4) and (A6)]. In formula (12g) the summation over index q is implied.

Equations (10) give an extinction theorem in the same form as in the case of a dense, $kb\to 0$, medium [see I, Eqs. (20)]. Furthermore, by inserting Eqs. (11) and (12) into Eq. (8) we obtain the relationship between the macroscopic field $\vec{E}$, which satisfies the wave equation (9a), and the microscopic (local) field $\vec{E}\,'$ acting on a given elementary radiator. A similar procedure is done for the magnetic field $\vec{H}$. In the routine case of a plane wave polarization this relationship takes a customary form,

$$\vec{E}=\vec{E}\,'+\hat{\beta}\cdot\vec{P}; \tag{13}$$

however, tensor $\hat{\beta}$, generally speaking, now depends on the wave vector $\vec{k}$ of the plane wave [see Eqs. (8) and (12)]. Therefore, the discreteness of a medium gives rise to a spatial dispersion of a certain type.

If one knows the microscopic properties of the elementary radiators, i.e., the relationship of polarization $\vec{P}$ to the local field $\vec{E}'$, then Eq. (13) allows us to obtain the macroscopic material equations. So, in the simplest case of a linear medium with the given polarizability tensor $\hat{\alpha}$ of a separate radiator we arrive at the well-known relation between microscopic $(\hat{\alpha},\vec{E}')$ and macroscopic $(\vec{P})$ quantities

$$\vec{P}=N\hat{\alpha}\cdot\vec{E}'. \tag{14}$$

As a result, Eqs. (9a), (13), and (14) give the dielectric permittivity tensor $\hat{\epsilon}$ with the allowance of a discreteness of the medium

$$\hat{\varepsilon}=1+4\pi N\hat{\alpha}\cdot(1+\hat{\beta}\cdot N\hat{\alpha})^{-1}, \tag{15a}$$

$$\hat{\beta}=\hat{\beta}_0+b^2\hat{\beta}_2:\vec{k}\vec{k}, \tag{15b}$$

where $\vec{k}$ is the wave vector of the propagating plane wave. In the limiting case, $kb\rightarrow 0$, Eqs. (15) are reduced to the former result [see I, Eq. (28)].

Now take notice of the nonlinear optics. The local field factor $\hat{f}_p$, which relates the right-hand part of the macroscopic wave equation with nonlinear polarization, may be obtained in the same manner as in a dense medium case I:

$$\hat{f}_p=(1+\hat{\beta}\cdot N\hat{\alpha})^{-1}. \tag{15c}$$

III. THE ROLE OF RADIATION DAMPING (RD)

Let us analyze Eqs. (15) taking into account the smallness of the parameter kb. It may be rewritten in the form

$$\hat{\varepsilon}-1=(\hat{\varepsilon}_0-1)\cdot\left[1-(kb)^2(\hat{\beta}_0^{(2)}+\hat{\beta}_2:\vec{q}\vec{q})\cdot\frac{\hat{\varepsilon}_0-1}{4\pi}\right]^{-1}-\frac{i}{6\pi}(kb)^3(\hat{\varepsilon}_0-1)^2, \tag{16a}$$

where $\vec{q}\equiv\vec{k}/k$ and the dielectric permittivity tensor $\hat{\varepsilon}_0$ in the limiting case, $k\rightarrow 0$, is given by

$$\hat{\varepsilon}_0-1=4\pi N\hat{\alpha}\cdot\hat{f}_{p0}. \tag{16b}$$

Here

$$\hat{f}_{p0}=(1+\hat{\beta}_0^{(0)}\cdot N\hat{\alpha})^{-1}=1-\hat{\beta}_0^{(0)}\cdot\frac{\hat{\varepsilon}_0-1}{4\pi}=\frac{\hat{\varepsilon}_0+2}{3}+\hat{\gamma}_0\cdot\frac{\hat{\varepsilon}_0-1}{4\pi} \tag{16c}$$

is the local field factor in this limit I, [5]. In the case of a cubic lattice or random media, $\hat{\gamma}_0=0$, one comes to the conventional LL limit.

The tensors $\hat{\beta}_0^{(0)}$ and $\hat{\beta}_0^{(2)}$ are given by the relations

$$\hat{\beta}_0^{(0)}=-\frac{4\pi}{3}-\hat{\gamma}_0, \tag{16d}$$

$$\hat{\beta}_0^{(2)}=\frac{8\pi}{15}(\hat{\Phi}-2\,\mathrm{Tr}\,\hat{\Phi})-\hat{\gamma}_0^{(2)}. \tag{16e}$$

Equation (16a) reveals the surprising fact that with an account of a medium's discreteness the dielectric permitivity (and, therefore, the refractive index) have *negative imaginary* parts, i.e., provide for amplification in a passive medium composed from the radiators having purely real microscopic polarizabilities $\hat{\alpha}$. The imaginary part $\hat{\varepsilon}''$ of the dielectric permittivity in Eq. (16a) may be rewritten in terms of the parameter Nb^3 as

$$\hat{\varepsilon}''=-\frac{8\pi}{3}N^2b^3k^3(\hat{\alpha}\cdot\hat{f}_p)^2. \tag{16f}$$

For any regular lattice the parameter Nb^3 is equal to unit due to the definition of the b, whereas for the gaslike medium this parameter is less then unit but may be fixed to unit for any medium by decreasing of a temperature. Therefore, this amplification does not relate directly to the value of the parameter kb so that the problem of violation of the energy conservation law exists also for *dense*, $kb\rightarrow 0$, media as well.

At first sight this seems paradoxical. To clarify the problem let us consider the limiting case of an ''ultrararefied,'' $kb\rightarrow\propto$, medium, i.e., the case of an isolated radiator. First, calculate the total flux of the electromagnetic energy from such a solitary dipole with a dipole moment $\vec{p}$

$$\vec{p}=\alpha\vec{E}, \tag{17a}$$

$$\alpha=\alpha'+i\alpha'', \tag{17b}$$

where microscopic polarizability α is an arbitrary scalar function of the field and frequency under irradiation of a plane monochromatic wave. The result is given by the flux of the Poynting vector $\vec{S}$

$$\vec{S}=\frac{c}{8\pi}\mathrm{Re}[\vec{E}\times\vec{H}], \tag{18a}$$

through a surface Σ of a sphere of an arbitrary radius around the dipole. A direct integration in the far-distant zone gives

$$\int_\Sigma\vec{S}\cdot\vec{n}_\Sigma d^2\vec{r}_\Sigma=\omega\left(\frac{k^3}{3}|\vec{p}|^2+\frac{1}{2}\,\mathrm{Im}\,\vec{p}\cdot\vec{E}^*\right)=\omega\left(\frac{k^3}{3}|\alpha|^2-\frac{\alpha''}{2}\right)|\vec{E}|^2. \tag{18b}$$

Here the first term corresponds to the radiation of the dipole, whereas the second term appears due to the interference of the dipole radiation with the incident field. (This result may be obtained in the course of more intricate calculations in any, not only far distant, zone and for any type of the monochromatic wave. In the last case $\vec{E}$ will be an amplitude of the electric field at the point of the dipole's disposition.)

Thus again the problem of energy conservation arises now for an isolated oscillator, since if one ascribes to a passive frictionless dipole the purely *real polarizability*, then the total flux of energy from such a dipole will never be zero. As long as a passive oscillator has no internal sources of energy we fall into a flagrant contradiction with the energy conservation law. Therefore, one must attribute to the polarizability of the oscillator the imaginary term α''

$$\alpha''=\frac{2}{3}\,k^3|\alpha|^2. \tag{18c}$$

One may consider it as a direct consequence of the optical theorem which connects the total cross section with the forward scattering amplitude. If we employ this formula to the model of a harmonic oscillator with a friction caused by the well-known radiation damping force f_{RD}

$$\vec{f}_{\mathrm{RD}}=-\frac{2}{3}\,q^2k^2\,\frac{\vec{\nu}}{c}\rightarrow\alpha \tag{18d}$$

$$=\alpha'+i\alpha''=\frac{q^2}{m}\,\frac{\omega_0^2-\omega^2+2i\omega\Gamma_{\mathrm{RD}}}{(\omega_0^2-\omega^2)^2+(2\omega\Gamma_{\mathrm{RD}})^2}, \tag{18e}$$

where $\vec{\nu}$ is the velocity of charge, ω_0 is the resonant frequency, and

$$\Gamma_{\mathrm{RD}}\equiv\frac{q^2k^2}{3mc}, \tag{18f}$$

then Eq. (18c) is satisfied identically. This result is a direct demonstration of the necessity of the RD account for a self-consistency of classical molecular optics. However, it has a broader sense. Really, Eq. (18a) was deduced without any concrete assumption about the internal structure of an oscillator. Therefore, it holds true for any type of oscillator, in particular for a nonlinear oscillator when the effects of the harmonics generation type may be neglected but the dependence of the polarizability α on the intensity $|\vec{E}|^2$ is still substantial (as in the case of self-focusing effects, etc.).

How are these results for an isolated oscillator related to the molecular optics approach in the broad sense and to the Lorentz-Lorenz formula in particular? From this direct electrodynamical consideration it is getting evident that even in the case of a ''frictionless'' but charged oscillator one must *always* take account of its losses. However, it must be done *not* in the form of substitution of the α'' term into the Lorentz-Lorenz formula, as it is done in some classical textbooks [12], since it leads to the appearance of the physically groundless ''intrinsic absorption'' in any homogeneous dielectric medium. Such a substitution should be done, in fact, into Eq. (16a). Then it meets the situation: in that way the propagation delay effects related to the phase difference between the neighboring oscillators is taking into account. In particular, for any regular lattice (when $Nb^3=1$) the effective ''amplification'' due to the collective interactions exactly cancels the RD losses. This Mandelstam's result for the regular isotropic medium (Mandelstam cancellation) is the main point of his basic consideration [9].

There is another way to solve the problem of the proper account of the RD effects in the framework of the molecular optics. We may *postulate* the necessity to take account of an oscillator's self-action. Mathematically it means extension of the summation over all oscillators, the proper one inclusive. The advantage of such an approach was demonstrated by Landau and Lifshitz [16] as applied to any charged system with sizes much less then the wavelength. They showed that for the calculation of the RD force it is sufficient to include the propagation delay effects into the ordinary Coulomb force $q\vec{E}$ acting on the charge q in the external field $\vec{E}$. It takes the form of the additional force $q\vec{E}_{\mathrm{RD}}$, where $\vec{E}_{\mathrm{RD}}$ is the first nondivergent term in the expansion of the delayed potential in terms of the parameter ν/c. In our technique it means that under calculation of the tensor $\hat{\gamma}_0$ (see Appendix B) for any geometry of the lattice one must expand the summation in Eq. (B3b) for the terms proportional to $i(kb)^3$ also to the proper ''l'' oscillator. Therefore, the term $\sim i(kb)^3$ in Eq. (12a) for local field factor $\hat{\beta}_0$ turns to zero and, as a result, the macroscopic and microscopic fields remain related by the purely real factor. This is a generalization of the Mandelstam cancellation effect in the case of an arbitrary anisotropic medium.

Summarizing, we have two equivalent methods for the calculation of the dielectric permittivity of a regular medium: (i) one may use Eq. (16a) and introduce into it the RD term α''_{RD} [see formula (18d)], or (ii) under calculation of the tensor $\hat{\gamma}_0$ one may extend the summation procedure over all radiators, the proper one inclusive. In this case the final result does not require any additional account of the RD effect, α does not acquire an imaginary part, and the dielectric permittivity remains real. Although both of these methods lead to the same result for dielectric permittivity, the second approach seems to us more suitable, in particular, for analysis of the nonlinear effects.

IV. THE OPTICAL PROPERTIES OF HIGHLY SYMMETRIC MEDIA

Generally speaking, any random spatial distribution of the particles is characterized by a set of the correlation functions describing the relative probabilities of their positions. Here we will emphasize a distinction between two cases of the centrosymmetric random structures—gaslike and jellylike media—as well as their characteristic difference from a regular cubic lattice.

A. Gaslike random medium

The simplest model for the description of random media with finite particle sizes is a model of ''hard spheres,'' which operates with two parameters—density N of the particles and a minimal possible distance b between them. In such a model the two-particle probability of finding particle ''j'' at any point of the volume, except the region $R_{jl}\leq b$ around the particle ''l,'' is constant, while inside this region this probability is equal to zero. This model has some relation to the case of a cooled trapped resonance gas, when due to the repulsive forces atoms cannot approach each other nearer than a certain distance [17]. For a cooled atomic gas the minimum distance b depends on temperature, detuning, intensity of a resonant field, etc.

A gaseous medium approximation implies that the condition

$$Nb^3 \ll 1 \tag{19}$$

is satisfied and so all multiparticle correlation functions may be factorized with an accuracy up to this parameter. After averaging over a statistical ensemble, the local field acting on the particle ''l'' is equal to the field of a continious infinite medium with a spherical cavity of radius b, centered at the point $\vec{r}_l$. Therefore, one may replace a sum by the integral, in this case without any additional splitting procedure [i.e., in Eq. (5) one ought to put terms $\vec{E}_b$ and $\vec{H}_b$ equal to zero]. So, in Eqs. (12) tensor $\hat{\Phi} \equiv 0$ and for the tensors $\hat{\gamma}$ we have formulas (B5a) and (B5b) of Appendix B. As a result, the relationships (8) between the local and macroscopic fields acquire the form

$$\vec{E} = \vec{E}' - \left\{ \frac{4\pi}{3} [1-(kb)^2] + \frac{2}{3} ik^3 \left(\frac{1}{N} - b^3 \right) \right\} \vec{P} + \frac{2\pi}{5} b^2 \left(\nabla\nabla\cdot\vec{P} - \frac{1}{3}\Delta\vec{P} \right), \tag{20a}$$

$$\vec{H} = \vec{H}' - \frac{2\pi}{3} ikb^2 \nabla\times\vec{P}. \tag{20b}$$

Here terms with the spatial derivatives correspond to a spatial dispersion arising due to a discreteness of the medium. For a linear isotropic medium ($\vec{P} = N\alpha\vec{E}'$) from Eq. (20a) and Maxwell equation $\nabla\cdot(\vec{E}+4\pi\vec{P})=0$ (see also Sec. VI) we obtain

$$\left\{ 1 + \frac{4\pi}{3} N\alpha[2+(kb)^2] + \frac{2}{3} ik^3\alpha(Nb^3-1) \right\} \nabla\cdot\vec{P} + \frac{4\pi}{15} N\alpha b^2 \Delta\nabla\cdot\vec{P} = 0, \tag{21a}$$

Since in a dense medium limiting case we have $\nabla\cdot\vec{P}=0$, then from Eq. (21a) it follows that

$$\nabla\cdot\vec{P} \sim N\alpha k^3 b^2 \vec{P}. \tag{21b}$$

Therefore, the term $b^2\nabla\nabla\cdot\vec{P}$ in Eq. (20a) is of an order of $(kb)^4$ and may be omitted. This fact allows us to find the refractive index n (dielectric constant ε) of a gas and the local field factor f_p:

$$\varepsilon - 1 = n^2 - 1 = (n_0^2-1)\left\{ 1 - \frac{n_0^2-1}{3}\left[\left(1+\frac{n_0^2}{10}\right)(kb)^2 + \frac{ik^3}{2\pi}\left(b^3 - \frac{1}{N}\right) \right] \right\}, \tag{22a}$$

$$f_p = \frac{n_0^2+2}{3}\left\{ 1 - \frac{n_0^2-1}{3}\left[\left(1+\frac{n_0^2}{10}\right)(kb)^2 + \frac{ik^3}{2\pi}\left(b^3 - \frac{1}{N}\right) \right] \right\}, \tag{22b}$$

where the refractive index n_0 without allowance for the delay effects ($k=0$) is given by the LL formula

$$n_0^2 - 1 = \frac{4\pi N\alpha}{1-(4\pi/3)N\alpha}. \tag{23}$$

The contribution of discreteness into the dielectric permittivity is proportional to the factor $(kb)^2$. The absence of the linear kb term is occured through the symmetric properties of a medium [see Eqs. (6)] and radial dependence of the dipole radiation [see Appendix B, Eqs. (B1a) and (B2a)]. Really, due to the factor $(1-ikR)e^{ikR}$ the expansions of the radiation fields have no linear terms. The first nonvanishing *odd* term $(kb)^3$ describes the RD effect. On the other hand, for the fixed values of the parameters b and N and a small value of the parameter $N\alpha$, the effect of a discreteness is proportional to $(N\alpha)^2$, just as it takes place for the collective effects in dense medium. The absence of the linear $N\alpha$ term is not accidental. It represents the fact that in the limit $\alpha\to 0$ the distinction between local and macroscopic fields disappears and, therefore, the difference $\varepsilon-1$ must tend to $4\pi N\alpha$. Note that Eqs. (22) have a sense only in the situation *with* a propagating wave, when such a wave really exists. In other words, it is a question of the validity of the Maxwell equations (and also wave equations). Because of the probable origin of a scattering in a discrete rarefied medium the invariability of the Maxwell equations is not so evident. Fortunately, it is possible to demonstrate that the Maxwell equations remain valid in this situation as well (see Sec. VI). Finally, the negative sign of the discrete term reveals the appearance of nonzero phases (i.e., delay effects) in the interaction of neighboring oscillators, which leads to the attenuation of the collective effects. One may expect that for large values of the parameter kb the allowance of the discreteness would change to a sign of $\varepsilon-1$, in particular, the appearance of a region of the effective anomalous dispersion [18]. The allowance of the $(kb)^3$ terms leads to the factor $(1-Nb^3)$ in Eq. (22a). For an ideal gas $Nb^3=0$ and we come to the result of Ref. [12], whereas for a lattice this factor turns to zero and one comes to the effective Mandelstam cancellation (now proved for any anisotropic medium). For the nonideal gas this factor is not equal to zero and so absorption of the medium depends on the interparticle interactions and the temperature (compare with [10,11] where such an absorption is expressed in terms of the medium's density fluctuations).

B. Jellylike random medium

The previous result is valid under the conditions of the inequality (19). For a jellylike random medium the corresponding condition is

$$Nb^3 \leqslant 1, \tag{24}$$

which means that we have a uniform angular distribution of the oscillators relative to each other and an average distance between two close spaced oscillators is comparable with the minimal distance b. Now a passage from a sum to the integral requires a special splitting procedure. To satisfy the random character of the distribution function we must average the tensor $\hat{\Phi}$ in Eqs. (12) over all orientations of a ''split cube'' to imitate the random lattice. As a result, the tensor $\hat{\Phi}$ acquires the following form (see Appendix A):

$$(\hat{\Phi})_{st}=\frac{\delta_{st}}{24}. \tag{25}$$

After calculations similar to those for the gaslike medium we come to the integral equations with modified terms $\vec{E}_\sigma$ and $\vec{H}_\sigma$ (see Appendix B) and the additional terms $\vec{E}_b$ and $\vec{H}_b$, which are now not equal to zero. As a result, we obtain the integral equations of the same form where, instead of the real scale parameter b, the renormalized effective parameter $\tilde{b}$

$$\tilde{b}=b/\sqrt{3}, \tag{26}$$

enters into all terms with the exception of the last term in Eq. (22a). Therefore, the magnitude of the scattering essentially depends on the concrete model of a liquid. Equations (20)–(22) remain true for jellylike media as well. However, due to renormalization (26) the spatial dispersion properties of these two ''differently'' random media with the same parameter kb will differ three times. A threefold decrease of a spatial dispersion effects for a jellylike medium may be clarified by the following qualitative consideration. For a random medium the influence of the discreteness is determined by a certain weighted average distance between two closed spaced oscillators. Since the upper limit $\sim N^{-1/3}$ of such an averaging in the case of gaslike medium is more then for a jellylike medium (the lower limit b is the same), a ''gas-weighted'' average is more then the ''jelly-weighted'' one. Therefore, the macroscopic properties of a jellylike medium would have more resemblance to the properties of a dense medium, i.e., to the spatial dispersionless LL formula.

C. A cubic lattice

A specificity of the regular distribution of oscillators we consider in the frameworks of a simple cubic lattice model. The relationships between macroscopic $\vec{E}$ and $\vec{H}$ and local $\vec{E}'$ and $\vec{H}'$ fields in this case follow from Eqs. (8), (12), and Appendixes A and B:

$$E_s=E_s'+\left[-\frac{4\pi}{3}+C_1(kb)^2\right]P_s+b^2\left[C_2(\nabla\nabla\cdot\vec{P})_s+C_3\Delta P_s+C_4\frac{\partial^2 P_s}{\partial x_s^2}\right], \tag{27a}$$

$$\vec{H}=\vec{H}'-\frac{1}{2}C_1 ikb^2\nabla\times\vec{P}. \tag{27b}$$

The numerical constants of an order of unity C_1, C_2, C_3, C_4 are given in Sec. IV.

The macroscopic properties of a cubic lattice differ from those of the random media not only quantitatively through the C factors, but mainly qualitatively. The last non-vector-covariant term in Eq. (27a) violates isotropy inherent to the random media. Consequently, the principal opportunity of the manifestation of the directions of the crystallographic axes of generally accepted isotropic cubic crystals in the macroscopic optical phenomena do arise. For a two-dimensional system of quantum dots, at $kb\sim 1$ the similar manifestation was observed recently [8].

For a lattice constituted from the linear isotropic oscillators the term $C_2b^2\nabla\nabla\cdot\vec{P}$ in Eq. (27a) may be omitted for the same reason as in case of a random medium. Then the relationship (27a) between microfields and macrofields acquires the form

$$E_s=E_s'+\left[-\frac{4\pi}{3}+C_1(kb)^2\right]P_s+b^2\left[C_3\Delta P_s+C_4\frac{\partial^2 P_s}{\partial x_s^2}\right]. \tag{28a}$$

In the case of $\vec{P}=N\alpha\vec{E}$ tensor $\hat{\varepsilon}$ in Eqs. (15) has a diagonal form $\varepsilon_{st}=\varepsilon_s\delta_{st}$. For the plane incident wave we have

$$\varepsilon_s-1=(n_0^2-1)\left\{1+\frac{n_0^2-1}{4\pi}[(C_3+C_4q_s^2|e_s|)n_0^2-C_1]\times(kb)^2\right\}, \tag{28b}$$

where $\vec{q}$ is the unit vector in the direction of propagation $\vec{q}\equiv\vec{k}/k$ and $\vec{e}$ is the unit vector of the polarization of light. And so a cubic lattice with allowance for kb terms has rather intricate properties. Under propagation along any of the crystallographic axes the term $q_s^2|e_s|$ equals zero, so that $\varepsilon_{xx}=\varepsilon_{yy}=\varepsilon_{zz}$ and the medium looks similar to the isotropic one. The same isotropy, $\varepsilon_{xx}=\varepsilon_{yy}=\varepsilon_{zz}$ take place under propagation of a wave along the diagonal of the cube. Furthermore, in the direction along a diagonal of a face, $k_x=k_y\neq k_z$, a cubic lattice looks similar to a medium with tensor components $\varepsilon_{xx}=\varepsilon_{yy}\neq\varepsilon_{zz}$, i.e., as an uniaxial crystal. Finally, in a general case, from the optical view point, a cubic lattice looks similar to a biaxial crystal.

V. THE ROLE OF DISCRETENESS IN ELECTRIC-QUADRUPOLE AND MAGNETIC-DIPOLE MEDIA

There are two reasons for a special consideration of such media: (i) First, although in linear optics electric-quadrupole and magnetic-dipole radiations are small compared with a electric-dipole, in nonlinear optics their contributions may be comparable or even more than the nonlinearity due to dipole's nonharmonicity (see, for example, [19]); (ii) second, in a number of cases impurities in composite materials [20] may be considered as elementary radiators and the MIE approach may be applied [21]. For a regular arrangement of the impurities, and when their sizes are comparable with the interimpurity distances, the multipolar moments of those impurities start to play an important role. The following considerations apply to both of these situations.

Starting from exact sum-type equations for local fields $\vec{E}'$ and $\vec{H}'$ acting on the elementary radiator at point $\vec{r}_l$ with the quadrupole moment $\hat{q}$ and magnetic-dipole moment $\vec{m}$ [see I, Eq. (4)]

$$\vec{E}'(\vec{r}_l)=\vec{E}_i(\vec{r}_l)+\sum_{j\neq l}[\nabla\times\nabla\times\vec{p}(\vec{r}_j)G(R_{jl})-\nabla\times\nabla\times\nabla\cdot\hat{q}(\vec{r}_j)G(R_{jl})+ik\nabla\times\vec{m}(\vec{r}_j)G(R_{jl})], \tag{29a}$$

$$\vec{H}'(\vec{r}_l)=\vec{H}_i(\vec{r}_l)+\sum_{j\neq l}[\nabla\times\nabla\times\vec{m}(\vec{r}_j)G(R_{jl})+ik\nabla\times\nabla\cdot\hat{q}(\vec{r}_j)G(R_{jl})-ik\nabla\times\vec{p}(\vec{r}_j)G(R_{jl})], \tag{29b}$$

and using the above-described splitting procedure (Appendix A) we come to the integral equations for local fields $\vec{E}'$ and $\vec{H}'$ with an allowance of the $(kb)^2$ terms

$$\vec{E}'(\vec{r})=\vec{E}_i(\vec{r})+\vec{E}_\sigma(\vec{r})+\vec{E}_b(\vec{r})+\int_\sigma^\Sigma(\nabla\times\nabla\times\vec{P}G-\nabla\times\nabla\times\nabla\cdot\hat{Q}G+ik\nabla\times\vec{M}G)d^3\vec{r}'-b^2\int_\Sigma(\hat{\Phi}:\vec{n}_\Sigma\nabla')\nabla\times\nabla\times\vec{P}G-\nabla\times\nabla\times\nabla\cdot\hat{Q}G+ik\nabla\times\vec{M}G)d^2\vec{r}_\Sigma, \tag{30a}$$

$$\vec{H}'(\vec{r})=\vec{H}_i(\vec{r})+\vec{H}_\sigma(\vec{r})+\vec{H}_b(\vec{r})+\int_\sigma^\Sigma(-ik\nabla\times\vec{P}G+ik\nabla\times\nabla\cdot\hat{Q}G+\nabla\times\nabla\times\vec{M}G)d^3\vec{r}'-b^2\int_\Sigma(\hat{\Phi}:\vec{n}_\Sigma\nabla')\times(-ik\nabla\times\vec{P}G+ik\nabla\times\nabla\cdot\hat{Q}G+\nabla\times\nabla\times\vec{M}G)\times d^2\vec{r}_\Sigma. \tag{30b}$$

Here $\hat{Q}=N\hat{q}$ and $\vec{M}=N\vec{m}$ are quadrupole and magnetic-dipole densities, respectively, $G(\vec{R}_{jl})=e^{ikR_{jl}}/R_{jl}$, and other notations are the same as in Eqs. (5) and (10). The additional ''discrete'' contributions of the LS exterior in the most general case may be written as

$$\vec{E}_b=b^2[k^2\hat{\gamma}_{b0}\cdot\vec{P}+\hat{\gamma}_{b2}\vdots(\nabla\nabla\vec{P})+k^2\hat{\xi}_{b1}\vdots(\nabla\hat{Q})+\hat{\xi}_{b3}\vdots:(\nabla\nabla\nabla\hat{Q})-ik\hat{\gamma}_{bM1}:(\nabla\vec{M})], \tag{31a}$$

$$\vec{H}_b=b^2[k^2\hat{\gamma}_{b0}\cdot\vec{M}+\hat{\gamma}_{b2}\vdots(\nabla\nabla\vec{M})+ik^3\hat{\gamma}_{bM0}:\hat{Q}+ik\hat{\xi}_{bM2}::(\nabla\nabla\hat{Q})+ik\hat{\gamma}_{bM1}:(\nabla\vec{P})]. \tag{31b}$$

Dimensionless tensors $\hat{\gamma}_b$ and $\hat{\xi}_b$ are determined by the lattice geometry and may be expressed in terms of the ''splitting tensor'' $\hat{\Phi}$ (see Appendix A). The contributions $\vec{E}_\sigma$ and $\vec{H}_\sigma$ of the radiators inside LS can be written as

$$\vec{E}_\sigma=\hat{\gamma}_0\cdot\vec{P}+b^2\hat{\gamma}_2\vdots(\nabla\nabla\vec{P})+\hat{\xi}_1\vdots(\nabla\hat{Q})+b^2\hat{\xi}_3\vdots:(\nabla\nabla\nabla\hat{Q})-ikb^2\hat{\gamma}_{M1}:(\nabla\vec{M}), \tag{32a}$$

$$\vec{H}_\sigma=\hat{\gamma}_0\cdot\vec{M}+b^2\hat{\gamma}_2\vdots(\nabla\nabla\vec{M})+ik\hat{\xi}_{M0}:\hat{Q}+ikb^2\hat{\xi}_{M2}::(\nabla\nabla\hat{Q})+ikb^2\hat{\gamma}_{M1}:(\nabla\vec{P}). \tag{32b}$$

For some types of lattice geometry the dimensionless tensors $\hat{\gamma}$ and $\hat{\xi}$ are calculated in Appendix B.

Note, that for a medium with not so sharply outlined (''blurred'') boundary the $\vec{P}$, $\hat{Q}$ and $\vec{M}$ densities on the Σ surface turn to zero (see I, Sec. II) so that the surface integrals in Eq. (30) vanish. However, we will conserve these integrals to remind that the consideration of discrete effects for the media with sharp boundaries requires a special treatment.

Now, employing a general idea of substitution of the variables (see Sec. II), we introduce the new variables $\vec{E}$ and $\vec{H}$ in the most general, consistent with the requirements of a symmetry, form

$$\vec{E}=\vec{E}'+\hat{\beta}_0\cdot\vec{P}+b^2\hat{\beta}_2\vdots(\nabla\nabla\vec{P})+\hat{\eta}_1\vdots(\nabla\hat{Q})+b^2\hat{\eta}_3\vdots:(\nabla\nabla\nabla\hat{Q})-ikb^2\hat{\beta}_{M1}:(\nabla\vec{M}), \tag{33a}$$

$$\vec{H}=\vec{H}+\hat{\beta}_0\cdot\vec{M}+b^2\hat{\beta}_2\vdots(\nabla\nabla\vec{M})+ik\hat{\eta}_{M0}:\hat{Q}+ikb^2\hat{\eta}_{M2}::(\nabla\nabla\hat{Q})+ikb^2\hat{\beta}_{M1}:(\nabla\vec{P}), \tag{33b}$$

where the tensors $\hat{\beta}$ and $\hat{\eta}$ are still free parameters, just as in the case of the tensors $\hat{\beta}$ in Eqs. (8). Then again, as in the case of the electric-dipole media, suppose that the new variables satisfy, in addition to Eqs. (30), the wave equations in the following form [see I, Eqs. (12)]:

$$\nabla\times\nabla\times\vec{E}-k^2\vec{E}=4\pi k^2\left(\vec{P}-\nabla\cdot\hat{Q}+\frac{i}{k}\nabla\times\vec{M}\right), \tag{34a}$$

$$\nabla\times\nabla\times\vec{H}-k^2\vec{H}=4\pi k^2\left(\vec{M}+\frac{i}{k}\nabla\times\nabla\cdot\hat{Q}-\frac{i}{k}\nabla\times\vec{P}\right). \tag{34b}$$

As was shown in Appendix C one may factor an operator $\nabla\times\nabla\times$ outside a sign of the integral. Then, similarly to the case of the electric-dipole medium, one can transform the volume integrals in Eqs. (30) into surface integrals and come to the equations of the same form as Eqs. (10), but now with additional, proportional to $\hat{Q},\vec{M}$ and their spatial derivatives, terms. Imposing the condition that each group of the terms in Eqs. (30) with the same spatial dependence must vanish independently, we define the values of the parameters $\hat{\eta}$ [parameters $\hat{\beta}$ are given by Eqs. (12)]

$$\hat{\eta}_1=\frac{8\pi}{5}\hat{\delta}_4+\hat{\xi}_1'-\hat{\xi}_1-(kb)^2\hat{\xi}_{b1}, \tag{35a}$$

$$\hat{\eta}_3=\hat{\xi}_3'-\hat{\xi}_3-\hat{\xi}_{b3}, \tag{35b}$$

$$\hat{\eta}_{M0}=-\hat{\xi}_{M0}-(kb)^2\hat{\xi}_{bM0}, \tag{35c}$$

$$\hat{\eta}_{M2}=\hat{\xi}_{M2}'-\hat{\xi}_{M2}-\hat{\xi}_{bM2}. \tag{35d}$$

Here

$$(\hat{\delta}_4)_{stpq}\equiv\delta_{sq}\delta_{tp}, \tag{35e}$$

$$\hat{\delta}_4\vdots\nabla\hat{Q}=\nabla\cdot\hat{Q}, \tag{35f}$$

$$\hat{\xi}_1' = -\frac{2\pi}{5}(ka)^2\hat{\delta}_4, \tag{36a}$$

$$\hat{\xi}_1' \vdots \nabla\hat{Q} = -\frac{2\pi}{5}(ka^2)\nabla\cdot\hat{Q}, \tag{36b}$$

$$(\hat{\xi}_3')_{stpqkm} = \frac{2\pi}{7}\left(\frac{a}{b}\right)^2\left(\frac{2}{5}\,\delta_{sm}\delta_{tk}\delta_{pq} - \delta_{st}\delta_{pk}\delta_{qm}\right), \tag{36c}$$

$$b^2\hat{\xi}_3' \vdots :(\nabla\nabla\nabla\hat{Q}) = \frac{2\pi}{7}a^2\left(\frac{2}{5}\,\Delta\nabla\cdot\hat{Q} - \nabla\nabla\cdot\nabla\cdot\hat{Q}\right), \tag{36d}$$

$$(\hat{\xi}_{M2}')_{stpqk} = \frac{2\pi}{5}\left(\frac{a}{b}\right)^2\varepsilon_{stk}\delta_{pq}, \tag{36e}$$

$$b^2\hat{\xi}_{M2}' :: (\nabla\nabla\hat{Q}) = \frac{2\pi}{5}a^2\nabla\times\nabla\cdot\hat{Q}, \tag{36f}$$

where ε_{stk} is an antisymmetric unit tensor of the third rank. Such a choice of the parameters $\hat{\beta}$ and $\hat{\eta}$ guarantees conversion of Eqs.-(10)-type relationships into an extinction theorem:

$$\vec{E}_i + \nabla\times\nabla\times\int_\Sigma\left\{\frac{1}{4\pi k^2}\left(\vec{E}\,\frac{\partial G}{\partial\nu} - G\,\frac{\partial\vec{E}}{\partial\nu} + G\vec{n}_\Sigma\nabla'\cdot\vec{E}\right) + G\left(\hat{Q}\cdot\vec{n}_\Sigma + \frac{i}{k}[\vec{M}\times\vec{n}_\Sigma]\right) - b^2(\hat{\Phi}:\vec{n}_\Sigma\nabla') \times\left(\vec{P}G - \nabla\cdot\hat{Q}G + \frac{i}{k}\nabla\times\vec{M}G\right)\right\}d^2\vec{r}_\Sigma = 0, \tag{37a}$$

$$\vec{H}_i + \nabla\times\nabla\times\int_\Sigma\left\{\frac{1}{4\pi k^2}\left(\vec{H}\,\frac{\partial G}{\partial\nu} - G\,\frac{\partial\vec{H}}{\partial\nu} + G\vec{n}_\Sigma\nabla'\cdot\vec{H}\right) - \frac{i}{k}[\vec{n}_\Sigma\cdot\hat{Q}\times\nabla' G] + G[\vec{n}_\Sigma\times\nabla'\hat{Q}] + G[\vec{P}\times\vec{n}_\Sigma] - b^2(\hat{\Phi}\vec{n}_\Sigma\nabla')\left(\vec{M}G + \frac{i}{k}\nabla\times\nabla\cdot\hat{Q}G - \frac{i}{k}\nabla\times\vec{P}G\right)\right\}d^2\vec{r}_\Sigma = 0. \tag{37b}$$

As was mentioned earlier, for media with the ‘‘blurred’’ boundaries the $\vec{P}$, $\vec{Q}$, and $\vec{M}$ surface densities vanish and an extinction theorem acquires the universal form of I, Eqs. (21). The expressions for the fields outside the medium (reflected waves) coincide, as usual, with the left-hand parts of Eqs. (37). Finally, by use of the Appendixes A and B, one may write the relationships between the macroscopic and microscopic fields for some types of media in an explicit form.

For a random medium we have

$$\vec{E} = \vec{E}' + \frac{4\pi}{3}[-1 + (k\tilde{b})^2]\vec{P} + \frac{2\pi}{5}\tilde{b}^2\left(\nabla\nabla\cdot\vec{P} - \frac{1}{3}\Delta\vec{P}\right) + \frac{2\pi}{5}[4 - (k\tilde{b})^2]\nabla\cdot\hat{Q} + \frac{2\pi}{35}\tilde{b}^2(2\Delta\nabla\cdot\hat{Q} - 5\nabla\nabla\cdot\nabla\cdot\hat{Q}) + \frac{2\pi}{3}ik\tilde{b}^2\nabla\times\vec{M}, \tag{38a}$$

$$\vec{H} = \vec{H}' + \frac{4\pi}{3}[-1 + (k\tilde{b})^2]\vec{M} + \frac{2\pi}{5}\tilde{b}^2\left(\nabla\nabla\cdot\vec{M} - \frac{1}{3}\Delta\vec{M}\right) + 2\pi ik\tilde{b}^2\left(\frac{1}{5}\nabla\times\nabla\cdot\hat{Q} - \frac{1}{3}\nabla\times\vec{P}\right), \tag{38b}$$

where $\tilde{b} = b$ for gaslike media and $\tilde{b} = b/\sqrt{3}$ for jellylike media. One can see the essential difference in local field correction $(kb)^2$ terms–a threefold decrease for a jellylike medium.

For a cubic lattice similar relations take the form

$$E_s = E_s' + \left[-\frac{4\pi}{3} + C_1(kb)^2\right]P_s + b^2\left[C_2(\nabla\nabla\cdot\vec{P})_s + C_3\Delta P_s + C_4\,\frac{\partial^2 P_s}{\partial x_s^2}\right] + \left[\frac{8\pi}{5} + 3g - C_2(kb)^2\right](\nabla\cdot\hat{Q})_s + \left[-\frac{15}{2}g_0 + \frac{1}{6}C_4(kb)^2\right]\frac{\partial Q_{SS}}{\partial x_s} + b^2\left\{C_5(\nabla\nabla\cdot\nabla\cdot\hat{Q})_s + C_6(\Delta\nabla\cdot\hat{Q})_s + C_7\,\frac{\partial}{\partial x_s}\left[\sum_t\frac{\partial^2 Q_{tt}}{\partial x_t^2} + 2\,\frac{\partial}{\partial x_s}(\nabla\cdot\hat{Q})_s + \Delta Q_{ss}\right] + C_8\sum_t\frac{\partial^3 Q_{ts}}{\partial x_s^3} + C_9\,\frac{\partial^3 Q_{ss}}{\partial x_s^3}\right\} - \frac{1}{2}C_1ikb^2\nabla\times\vec{M}, \tag{39a}$$

$$H_s = H_s' + \left[-\frac{4\pi}{3} + C_1(kb)^2\right]M_s + b^2\left[C_2(\nabla\nabla\vec{M}_s) + C_3\Delta M_s + C_4\,\frac{\partial^2 M_s}{\partial x_s^2}\right] + \frac{1}{2}C_1ikb^2\nabla\times\vec{P} + C_2ikb^2(\nabla\times\nabla\cdot\hat{Q})_s + C_4\sum_{p,k}\varepsilon_{spk}\,\frac{\partial^2 Q_{pk}}{\partial x_p^2}, \tag{39b}$$

where the numerical calculations of C resulting in

$$C_1 = \frac{2g_0 + \pi/3}{3} = 1.892, \tag{40a}$$

$$C_2 = \frac{1}{2}\left(g_0 - 3g_1 + \frac{\pi}{15}\right) = 1.237, \tag{40b}$$

$$C_3 = \frac{1}{4}\left(\frac{1}{3}g_0 - 3g_1 - \frac{\pi}{45}\right) = 0.059, \tag{40c}$$

$$C_4=\frac{3}{4}\,(5g_1-g_0)=-1.675, \tag{40d}$$

$$C_5=5\left(\frac{9}{2}\,g_1-\frac{g_0}{2}-3g_2\right)-\frac{\pi}{42}=21.204, \tag{40e}$$

$$C_6=21g_1-2g_0-15g_2+\frac{\pi}{105}=22.442, \tag{40f}$$

$$C_7=\frac{15}{2}\left(\frac{g_0}{2}-5g_1+\frac{7}{2}\,g_2\right)=-38.655, \tag{40g}$$

$$C_8=2g_0-\frac{45}{2}\,g_1+\frac{35}{2}\,g_2=-26.886, \tag{40h}$$

$$C_9=\frac{1}{2}\left(\frac{105}{2}\,g_1-5g_0-\frac{77}{2}\,g_2\right)=28.905, \tag{40i}$$

where g (see I), g_0, g_1, and g_2 are the lattice sums:

$$g=\lim_{A\to\infty}\sum_{m^2+n^2+k^2\neq 0}^{m^2+n^2+k^2\leqslant A^2}\left[\frac{5m^4}{(m^2+n^2+k^2)^{5/2}}-\frac{1}{(m^2+n^2+k^2)^{3/2}}\right]$$
$$=3.113, \tag{41a}$$

$$g_0=\lim_{A\to\infty}\left[2\pi A^2-\sum_{m^2+n^2+k^2\neq 0}^{m^2+n^2+k^2\leqslant A^2}(m^2+m^2+k^2)^{-1/2}\right], \tag{41b}$$

$$g_1=\lim_{A\to\infty}\left[\frac{2\pi}{5}\,A^2-\sum_{m^2+n^2+A^2\neq 0}^{m^2+n^2+k^2\leqslant A^2}\frac{m^4}{(m^2+n^2+k^2)^{5/2}}\right], \tag{41c}$$

$$g_2=\lim_{A\to\infty}\left[\frac{2\pi}{7}\,A^2-\sum_{m^2+n^2+k^2\neq 0}^{m^2+n^2+k^2\leqslant A^2}\frac{m^6}{(m^2+n^2+k^2)^{7/2}}\right]. \tag{41d}$$

Under numerical calculations of g_0, g_1, and g_2 with any given accuracy there is a problem of the right choice of the LS sphere radius: depending on the value of this radius the number of the boundary points, which is situated exactly on the LS surface and may be arbitrary considered as external or internal ones, increases and their contribution to the sums grows. This leads to the fictitious, depending on the concrete choice of the LS radius, ‘‘fluctuations’’ of the results. To overcome this difficulty we assumed the procedure of an averaging of this choice, i.e., at any given LS radius a certain ‘‘fluctuation’’ of the radius was introduced and the results were averaged. After such an averaging the results become independent of the value of the radius.

At the same time, we elaborated another procedure for calculation of the sums. It consists of a modification of the Lorentz-Cavity form. For a cubic lattice it is natural to take the cubic, instead of the spherical, form of a cavity. This approach leads to relationships which are equivalent to Eqs. (39) with the following modifications in Eqs. (40) (see Appendix E):

$$g_0=g_0'=2.313, \tag{42a}$$

$$g_1=g_1'+\frac{1}{36}\left(\frac{1409}{30}\,\pi-\frac{259}{\sqrt{3}}\right)=0.0161, \tag{42b}$$

$$g_2=g_2'+\frac{1}{72}\left(\frac{8309}{\sqrt{3}}-\frac{15629}{10}\,\pi\right)=-1.780, \tag{42c}$$

where

$$g_0'=\lim_{A\to\infty}\left\{2[6\ln(2+\sqrt{3})-\pi]\left(A+\frac{1}{2}\right)^2-\sum_{m,n,k,=-A}^{m,n,k=A}\frac{1}{(m^2+n^2+k^2)^{1/2}}\right\}=2.314, \tag{42d}$$

$$g_1'=\lim_{A\to\infty}\left\{2\left[2\ln(2+\sqrt{3})-\frac{5}{9}\,\pi\right]\left(A+\frac{1}{2}\right)^2-\sum_{m,n,k=-A}^{m,n,k=A}\frac{m^4}{(m^2+n^2+k^2)^{5/2}}\right\}=0.0712, \tag{42e}$$

$$g_2'=\lim_{A\to\infty}\left\{2\left[2\ln(2+\sqrt{3})-\frac{1}{5}\left(3\pi+\frac{4}{3\sqrt{3}}\right)\right]\left(A+\frac{1}{2}\right)^2-\sum_{m,n,k,=-K}^{m,n,k=A}\frac{m^6}{(m^2+n^2+k^2)^{7/2}}\right\}=-0.213. \tag{42f}$$

Formulas (33)–(36) and (38)–(42) give a solution of the local field problem. Now it is time to write the microscopic material equations connecting the densities $\vec{P}$, $\hat{Q}$, and $\vec{M}$ to the microscopic fields $\vec{E}'$ and $\vec{H}$. This is a problem of the microscopic theory of an elementary radiator. As a result, one can express all the values in the wave the Eqs. (34) through the macroscopic fields $\vec{E}$ and $\vec{H}$ and obtain completely self-consistent description of any optical phenomena.

VI. MAXWELL EQUATIONS

We formulate the last problem as follows. How, does the discreteness of the medium influence the form of Maxwell equations in this medium? For too ‘‘highly rarefied’’ media, under $kb \gg 1$, this problem appears unessential since, evidently, the equations must tend to those in vacuum. But what are the modifications for the case $kb \ll 1$, when kb is a finite parameter although small? To answer this question we start from the integral equations for microscopic fields $\vec{E}'$ and $\vec{H}'$ and go over, by use of Eqs. (12), (35), and (36), to integral equations for the macroscopic fields $\vec{E}$ and $\vec{H}$ in the form

$$\vec{E}=\vec{E}_i+\frac{4\pi}{3}\left[-1+(ka)^2+\frac{2}{3}\,i(ka)^3\right]\vec{P}+\frac{2\pi}{5}\,a^2\left(\nabla\nabla\cdot\vec{P}-\frac{1}{3}\,\Delta\vec{P}\right)+\frac{2\pi}{5}\,[4-(ka)^2]\nabla\cdot\hat{Q}+\frac{2\pi}{35}\,a^2(2\Delta\nabla\cdot\hat{Q}$$

$$-5\nabla\nabla\cdot\nabla\cdot\hat{Q})+\frac{2\pi}{3}ika^2\nabla\times\vec{M}+\int_\sigma^\Sigma(\nabla\times\nabla\times\vec{P}G$$
$$-\nabla$$
$$\times\nabla\times\nabla\cdot\hat{Q}G+ik\nabla\times\vec{M}G)d^3\vec{r}\,'-b^2\int_\Sigma(\hat{\Phi}:\vec{n}_\Sigma\nabla')$$
$$\times(\nabla\times\nabla\times\vec{P}G-\nabla\times\nabla\hat{Q}G+ik\nabla\times\vec{M}G)d^2\vec{r}_\Sigma, \tag{43a}$$

$$\vec{H}=\vec{H}_i+\frac{4\pi}{3}\left[-1+(ka)^2+\frac{2}{3}i(ka)^3\right]\vec{M}+\frac{2\pi}{5}a^2\bigg(\nabla\nabla\cdot\vec{M}$$
$$-\frac{1}{3}\Delta\vec{M}\bigg)+2\pi ika^2\left(\frac{1}{5}\nabla\times\nabla\cdot\hat{Q}-\frac{1}{3}\nabla\times\vec{P}\right)$$
$$+\int_\sigma^\Sigma(-ik\nabla\times\vec{P}G-ik\nabla\times\nabla\cdot\hat{Q}G+\nabla\times\nabla\times\vec{M}G)d^3\vec{r}\,'$$
$$-b^2\int_\Sigma(\hat{\Phi}:\vec{n}_\Sigma\nabla')(-ik\nabla\times\vec{P}G+ik\nabla\times\nabla\cdot\hat{Q}G+\nabla\times\nabla$$
$$\times\vec{M}G)d^2\vec{r}_\Sigma. \tag{43b}$$

Now we apply the operators div and rot to the left-hand and right-hand parts of Eqs. (43). As shown in Appendix C, these operators can be insert under the integrals. The resulting expressions must be compared with Eqs. (43). Finally, we come to the ordinary macroscopic Maxwell equations *without any correction of the form* due to the discreteness of the medium:

$$\nabla\cdot(\vec{E}+4\pi\vec{P}-4\pi\nabla\cdot\hat{Q})=\nabla\cdot\vec{D}=0, \tag{44a}$$

$$\nabla\times\vec{E}=ik(\vec{H}+4\pi\vec{M})=ik\vec{B}=-\frac{1}{c}\frac{\partial\vec{B}}{\partial t}, \tag{44b}$$

$$\nabla\cdot(\vec{H}+4\pi\vec{M})=\nabla\cdot\vec{B}=0, \tag{44c}$$

$$\nabla\times\vec{H}=-ik(\vec{E}+4\pi\vec{P}-4\pi\nabla\cdot\hat{Q})=-ik\vec{D}=\frac{1}{c}\frac{\partial\vec{D}}{\partial t}, \tag{44d}$$

where the definitions of quantities $\vec{D}$ and $\vec{B}$ are

$$\vec{D}\equiv\vec{E}+4\pi\vec{P}-4\pi\nabla\cdot\hat{Q}, \tag{45a}$$

$$\vec{B}\equiv\vec{H}+4\pi\vec{M}. \tag{45b}$$

In this way, the allowance of the medium's discreteness with an accuracy to $(kb)^3$ terms, inclusive, not at all influence the form of the Maxwell equations. It is by no means a self-evident result. The proved preservation of the Maxwell and wave equations in rarefied medium with an accuracy up to the third order in the parameter kb signifies the absence of the scattering at least with the same accuracy. It gives the basis to presume that an origin of the scattering in a regular discrete medium would take place in the threshold manner.

VII. SUMMARY AND DISCUSSION

A special procedure of the ''radiator splitting'' in the case of a weakly rarefied media allows us to pass from the exact sum-equations of molecular optics [see Eqs. (29) and I, Eqs. (4)] to the integral equations and so it is of a major importance in all these considerations. The general idea of a substitution of the variables into the integral equations, which corresponds to the passage from the acting $(\vec{E}\,',\vec{H}\,')$ to the macroscopic $(\vec{E},\vec{H})$ fields, was found suitable not only for highly condensed matter $(kb\rightarrow 0)$ but also for the weakly rarefied media.

To outline the results let us separate the problems of the macroscopic properties of an optical medium into two groups: (i) the case of a medium with regular internal structure (an ideal crystalline lattice at zero temperature) and (ii) the case of an irregular medium (gaslike or jellylike media and imperfect crystals or crystals at nonzero temperature).

(i) In this case a self-consistent theory is developed and the results can be summarized as follows.

(1) If, in accordance with the general concept, one would account for the RD effects by means of the formal substitution of the microscopic polarizability $\alpha=\alpha'+\alpha''$ of an isolated radiator into the LL relation [12], one arrives at the erroneous conclusion about specific fictitious damping inherent to any dielectric medium. It is going on due to the neglection of the propagation delay effects (phase difference) in the interaction of the neighbor radiators, related to the effective amplification. Both of these effects are exactly balanced against each other. For the particular case of a homogeneous isotropic medium it was shown first by Mandelstam [9] (Mandelstam cancellation), although not quite rigorously (he ignored two important factors—the spatial dispersion inside the LS and the medium's discreteness outside it—which did not influence the final conclusion only due to specific symmetric properties of the isotropic medium). This is the reason why the Mandelstam cancellation effects do not have universal character and Planck's argumentation is also relevant, at least relative to a random medium. For the regular media the validity of the Mandelstam cancellation effects is proved up to any order in the parameter kb, up to the origin of the Bragg diffraction [14].

(2) It is shown that the physical notion of the macroscopic fields $\vec{E}$ and $\vec{H}$ may be deduced without any averaging proceeding straightforwardly from the formal mathematical properties of the initial integral equations.

(3) The extinction theorem is deduced. For the medium with a ''blurred'' boundary it has an identical form to the case of highly condensed matter.

(4) We demonstrated, to an accuracy up to $(kb)^3$ terms, that for a weakly rarefied medium macroscopic Maxwell equations keep a generally accepted form.

(5) The $(kb)^2$ corrections to the dielectric permittivity tensor $\hat{\varepsilon}$ and local field factors arising due to the finite value of the parameter kb were calculated. Depending on this microscopic parameter there are the rarefied media with the essentially different macroscopic properties. This conclusion refers to a gaslike, jellylike, and a cubic lattice media. The spatial dispersion of such media is calculated. For a cubic lattice crystal an optical anisotropy is revealed.

(ii) In the case of a random distribution of the elementary radiators or with allowance of the thermodynamic fluctuations one should calculate the local fields for a given spatial distribution of the dipole moments and only after that perform the averaging over an ensemble. In this paper we operate, in fact, in the other way: we *initially* average the multipole moments and positions of each particle over all possible states and only afterwards calculate the acting fields. It is clear that such a procedure is true only in the case of sufficiently rapid processes of ''self-averaging'' over internal and external degrees of freedom of each particle. This situation is definitely realized, for example, in a dense (in sense of the condition $N\lambda^3 \gg 1$) medium and for an elastic mechanism of polarizability. On this assumption it is possible to show the validity of the extinction theorem, Maxwell equations, and calculate the macroscopic optical properties of the medium. It is interesting that even an elementary model of a weakly rarefied medium demonstates not only quantitative properties—such as the modification of the LL formula—but also some qualitative characteristics which are evident from the example of gaslike and jellylike media.

When the problem of local field factors is solved, the influence of fluctuations inside of the Lorentz cavity must be taken into account. In the case of the elastic mechanism of polarizability their contribution takes the form of corrections, whereas in some other cases, for example, for the orientational mechanizm of the polarizability, they play a predominant role. Earlier, for condensed matter, the allowance of the fluctuations was made in the form of renormalization of only the real part of the polarizability, so that the microfields and macrofields remained related by a pure real factor (see, for example, [22]). Now we see that when the discreteness of a medium is properly taken into account the fluctuations lead to an appearance of the imaginary part of this factor. Therefore, an absorption would arise in the fluctuating medium, which otherwise would be considered as a transparent one. As is evident, the physical origin of the absorption is scattering of the light. The developed approach permits calculation of the integral intensity of scattering, depending on the parameter kb. It follows from Eq. (12a) and the Appendix B that the imaginary term $\sim(kb)^3$ in the local field factors will be determined by the fluctuations of the total dipole moment of the LS. When the complex refractive index is calculated, and hence the spatial distribution of the incident wave and polarization $\vec{P}$ is known, then one can calculate the angular distribution of the scattered radiation with the help of the integrals in the right-hand parts of Eq. (5) or Eq. (30).

As is known, in the case of a regular medium the appearance of the Bragg scattering takes place only after the parameter kb exceeds a certain value. Our results for a weakly rarefied medium may be considered as the first step to the analytic description of the photonic band gap structures. For an irregular medium a scattering effect occurs in a gradual manner [23]. This fact justifies, to some extent, an approach using the direct substitution [12] of the RD (imaginary part of the polarizability) into the LL formula under consideration of irregular (but in no circumstances regular) media. It is groundless, of course, to expect exact numerical agreement with the experimental data in all cases with the important exception of the ideal gas medium [10,11]. The present theory has no difficulty in explaining the irregular fluctuating media: this problem may be treated with the help of the present approach which may be complemented by the generalization of the Frohlich theorem for an isotropic medium [24] to the case of the anisotropic media [25].

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APPENDIX A: THE CALCULATION OF THE DIFFERENCE BETWEEN INTEGRAL AND SUM OUTSIDE THE LS

The electric or magnetic fields (here designated as $\vec{U}$) from any electric-dipole, electric-quadrupole, or magnetic-dipole radiator may be written in the form

$$\vec{U}(\vec{r}_l)=\hat{L}(\nabla_l)G(\vec{r}_j-\vec{r}_l)\hat{f}(\vec{r}_j)\,, \tag{A1}$$

where $\hat{L}(\nabla_l)$ is a definite differential operator acting on $\vec{r}_l$ coordinates of the LS center and $\hat{f}$ is a tensor or vector of the volume densities of the electric-dipole, electric-quadrupole, or magnetic-dipole moments. Let us split, mentally, each of the elementary radiators into eight smaller ones with the same total multipole moment as an initial ''unsplit'' radiator. The displacements of the new elementary radiators must be chosen in such a way that while the geometry of the new lattice would remain intact, its lattice constant would reduce by a factor of 2 (see Fig. 1). Then, using the inequality

$$R_{jl}\gg a\gg b, \tag{A2}$$

one can calculate the field in the LS center. For this it is necessary to substitute into Eq. (A1) the vector $\vec{r}_j+\vec{b}_{j'}$ and expand this expression into Taylor series of $\vec{b}_{j'}$:

$$\vec{U}=\hat{L}G\hat{f}+\frac{1}{8}\frac{\partial}{\partial(\vec{r}_j)_s}\hat{L}G\hat{f}\sum_{j'}(\vec{b}_{j'})_s+\frac{1}{16}\frac{\partial^2}{\partial(\vec{r}_j)_s\partial(\vec{r}_j)_t}\times\hat{L}G\hat{f}\sum_{j'}(\vec{b}_{j'})_s(\vec{b}_{j'})_t+\cdots\,. \tag{A3}$$

Here j' takes on values from 1 to 8 and $\vec{r}_j+\vec{b}_{j'}$ are radius vectors of the splited oscillators. In Eq. (A3) the summation is implied over all indices s and t. As known, any lattice constituted from identical radiators has a center of symmetry and due to this fact the second term in the right-hand part of Eq. (A3) turns to zero. Therefore, performing the summation over j in both of the parts (A3), one obtains

$$\sum_j \hat{L}G\hat{f}=\frac{1}{8}\sum_j {}^{(1)}\hat{L}G\hat{f} -\frac{1}{8}b^2\sum_j {}^{(1)}\Phi'_{st}\frac{\partial^2}{\partial(\vec{r}_j)_s\partial(\vec{r}_j)_t}\hat{L}G\hat{f}, \tag{A4a}$$

where

$$(\hat{\Phi}')_{st}\equiv\frac{1}{16b^2}\sum_{j'}(\vec{b}_{j'})_s(\vec{b}_{j'})_t \tag{A4b}$$

and $\Sigma_j^{(1)}$ denotes the summation over the new lattice. By reiterating this procedure N times we come to the conclusion that the difference between an initial sum and a sum with the $b/2^N$ period is equal to a geometrical progression, each term of which is proportional to b^2. This progression may be calculated directly, and as a result, we obtain an explicit expression for the difference between the initial sum and such a sum in the limiting case $kb\to0$, i.e., the integral over the LS exterior

$$\sum_{j\neq l}\hat{L}G\hat{f}=\int_\sigma^\Sigma[\hat{L}G\hat{F}-b^2(\hat{\Phi}:\nabla'\nabla')\hat{L}G\hat{F}]d^3\vec{r}', \tag{A5a}$$

where $\hat{F}=\hat{f}/b^3$ is the volume density of the quantity $\hat{f},\hat{\Phi}\equiv(4/3)\hat{\Phi}'$.

Owing to the presence of the operator ∇', a second volume integral may be transformed into the surface integrals over the outer boundary Σ and the LS surface σ. The latter can be calculated directly and gives rise to the terms $\vec{E}_b$ and $\vec{H}_b$ [see Eqs. (5) and (30)]. By applying this procedure to electric-dipole, electric-quadrupole, and magnetic-dipole media we come to the integral equations (5) and (30) with the following tensors $\hat{\gamma}_b$:

$$\hat{\gamma}_{b0}=\frac{8\pi}{15}(2\mathrm{Tr}\hat{\Phi}-\hat{\Phi}), \tag{A5b}$$

$$(\hat{\gamma}_{b2})_{stpq}=\frac{8\pi}{105}[6(\hat{\Phi})_{st}\delta_{pq}+3(\hat{\Phi})_{sq}\delta_{tp}-4(\hat{\Phi})_{tp}\delta_{sq}+6(\hat{\Phi})_{pq}\delta_{st}]+\frac{4\pi}{35}(\delta_{sq}\delta_{tp}-5\delta_{st}\delta_{pq})\mathrm{Tr}\hat{\Phi}, \tag{A5c}$$

$$(\gamma_{bM1})_{stp}=\frac{4\pi}{5}\left[\varepsilon_{stp}\mathrm{Tr}\hat{\Phi}-\frac{4}{3}\varepsilon_{stq}(\hat{\Phi})_{qp}\right]. \tag{A5d}$$

Due to rather an intricate character of the expressions for tensors $\hat{\xi}_b$ we present them only for the case of a cubic lattice (jellylike medium)—see Eq. (A8).

In the case of an orthorombic lattice from Eq. (A4) it follows

$$(\hat{\Phi})_{st}=\Phi_s\delta_{st}, \tag{A6a}$$

$$\Phi_s\equiv\frac{1}{12b^2}\sum_{j'}(\vec{b}_{j'})^2{}_s, \tag{A6b}$$

whereas for a cubic lattice we obtain

$$(\hat{\Phi})_{st}=\frac{1}{24}\delta_{st}. \tag{A7}$$

Tensors $\hat{\gamma}_b$ and $\hat{\xi}_b$ take the form

$$(\hat{\gamma}_{b0})_{st}=-\frac{\pi}{9}\delta_{st}, \tag{A8a}$$

$$\hat{\gamma}_{b0}\cdot\vec{P}=-\frac{\pi}{9}\vec{P}, \tag{A8b}$$

$$(\hat{\gamma}_{b2})_{stpq}=\frac{\pi}{30}\left(\frac{1}{3}\delta_{sq}\delta_{pt}-\delta_{st}\delta_{pq}\right), \tag{A8c}$$

$$\hat{\gamma}_{b2}:(\nabla\nabla\vec{P})=\frac{\pi}{30}\left(\frac{1}{3}\Delta\vec{P}-\nabla\nabla\cdot\vec{P}\right), \tag{A8d}$$

$$(\hat{\xi}_{b1})_{stpq}=\frac{\pi}{30}\delta_{sq}\delta_{tp}, \tag{A8e}$$

$$\hat{\xi}_{b1}\vdots(\nabla\hat{Q})=\frac{\pi}{30}\nabla\cdot\hat{Q}, \tag{A8f}$$

$$(\hat{\xi}_{b3})_{stpqkl}=\frac{\pi}{42}\left(\delta_{st}\delta_{pk}\delta_{ql}-\frac{2}{5}\delta_{sl}\delta_{tp}\delta_{qk}\right), \tag{A8g}$$

$$\hat{\xi}_{b3}\vdots:(\nabla\nabla\nabla\hat{Q})=\frac{\pi}{42}\left(\nabla\nabla\cdot\nabla\cdot\hat{Q}-\frac{2}{5}\Delta\nabla\cdot\hat{Q}\right), \tag{A8h}$$

$$(\hat{\gamma}_{bM1})_{stp}=\frac{\pi}{18}\varepsilon_{stp}, \tag{A8i}$$

$$\hat{\gamma}_{bM1}:(\nabla\vec{M})=\frac{\pi}{18}\nabla\times\vec{M}, \tag{A8j}$$

$$(\hat{\xi}_{bM2})_{stpqk}=-\frac{\pi}{30}\delta_{pq}\varepsilon_{stk}, \tag{A8k}$$

$$\hat{\xi}_{bM2}::(\nabla\nabla\hat{Q})=-\frac{\pi}{30}\nabla\times\nabla\cdot\hat{Q}, \tag{A8l}$$

$$\hat{\xi}_{bM0}:\hat{Q}=0. \tag{A8m}$$

For a gaslike random medium in the first order approximation in a gas parameter Nb^3 we have the same probability of finding a particle at any point of the space. The sole exception is a sphere of the radius b around the given radiator where this probability equal zero. After an ensemble averaging the medium looks, from a viewpoint of a given radiator, similar to a continuous one. Therefore, summation may be replaced by an integration without any ''splitting'' procedure at all, and the tensor $\hat{\Phi}$ must be equated to zero. Finally, for a jellylike medium we have an uniform angular distribution of the particles, but the distance between any two neighboring particles is approximately equal to b (unlike a gaslike medium where this distance may be as large as $N^{-1/3} \gg b$). It is evident that to average Eq. (A7) over the orientations one should rotate each of the splitted cell in an uniform way. Owing to such an averaging, the tensor $\hat{\Phi}$ does not depend on the orientation, i.e. it turns into a scalar which must be, with the b^2 accuracy, the same as for a cubic lattice.

APPENDIX B: CALCULATION OF THE TENSORS $\hat{\gamma}$ AND $\hat{\xi}$

Since we are breaking down with the $kb \to 0$ approximation, we must start from the explicit expressions for the electric $\vec{E}_p$, $\vec{E}_q$, $\vec{E}_m$ and magnetic $\vec{H}_p$, $\vec{H}_q$, $\vec{H}_m$ fields of dipole (p), quadrupole (q), and magnetic-dipole (m) elementary radiators:

$$\vec{E}_p = \nabla \times \nabla \times \vec{p}\, \frac{e^{ikR}}{R} = e^{ikR}\left\{ \frac{1-ikR}{R^3}[3\vec{n}(\vec{n}\cdot\vec{p})-\vec{p}] + \frac{k^2}{R}[\vec{p}-\vec{n}(\vec{n}\cdot\vec{p})]\right\}, \tag{B1a}$$

$$\vec{E}_q = \nabla \times \nabla \times \nabla \cdot \hat{q}\, \frac{e^{ikR}}{R} = e^{ikR}\left\{ 3\frac{1-ikR}{R^4}[5\vec{n}(\vec{n}\cdot\vec{n}\cdot\hat{q})-2\vec{n}\cdot\hat{q}] + 3\frac{k^2}{R^2}[\vec{n}\cdot\hat{q}-2\vec{n}(\vec{n}\cdot\vec{n}\cdot\hat{q})] + \frac{ik^3}{R}[\vec{n}(\vec{n}\cdot\vec{n}\cdot\hat{q})-\vec{n}\cdot\hat{q}]\right\}, \tag{B1b}$$

$$\vec{E}_m = ik\nabla \times \vec{m}\, \frac{e^{ikR}}{R} = -ike^{ikR}\frac{1-ikR}{R^3}\, \vec{n}\times\vec{m}, \tag{B1c}$$

$$\vec{H}_p = -ik\nabla \times \vec{p}\, \frac{e^{ikR}}{R} = ike^{ikR}\frac{1-ikR}{R^2}\, \vec{n}\times\vec{p}, \tag{B2a}$$

$$\vec{H}_q = -ik\nabla \times \nabla \cdot \hat{q}\, \frac{e^{ikR}}{R} = ike^{ikR}\frac{3(1-ikR)-(kR)^2}{R^3}\, \vec{n}\times(\vec{n}\cdot\hat{q}), \tag{B2b}$$

$$\vec{H}_m = \nabla \times \nabla \times \vec{m}\, \frac{e^{ikR}}{R} = e^{ikR}\left\{ \frac{1-ikR}{R^3}[3\vec{n}(\vec{n}\cdot\vec{m})-\vec{m}] + \frac{k^2}{R}[\vec{m}-\vec{n}(\vec{n}\cdot\vec{m})]\right\}, \tag{B2c}$$

where

$$\vec{R} \equiv \vec{R}_{jl} = \vec{r}_j - \vec{r}_l, \quad \vec{n} \equiv \vec{R}/R.$$

Using the same method as in paper I, Appendix A, but keeping now the $(kb)^3$ terms one may find the tensors $\hat{\gamma}, \hat{\xi}$ for a lattice of any kind. Further, we again take into account the existence of a center of symmetry of the medium so that any lattice sum constructed from terms with odd number of factors of the components of the unit vector $\vec{n}$ turns to zero. As a result, we have

$$(\hat{\gamma}_1)_{stp} = (\hat{\gamma}_3)_{stpqk} = (\hat{\xi}_0)_{stp} = (\hat{\xi}_2)_{stpqkm} = (\hat{\gamma}_M)_{st} = (\hat{\gamma}_{M2})_{stpq} = 0, \tag{B3a}$$

$$(\hat{\gamma}_0)_{st} = b^3 \sum \left(\frac{3n_s n_t - \delta_{st}}{R^3} + k^2\, \frac{\delta_{st} - n_s n_t}{R} \right) + \frac{2}{3}\, i(kb)^3 (N_\sigma - 1), \tag{B3b}$$

$$(\hat{\gamma}_2)_{stpq} = \frac{b}{2} \sum \frac{3n_s n_t n_p n_q - n_t n_p \delta_{sq}}{R}, \tag{B3c}$$

$$(\hat{\xi}_1)_{stpq} = 3b^3 \sum \left(\frac{2n_t n_p \delta_{sq} - 5n_s n_t n_p n_q}{R^3} - \frac{k^2}{2}\, n_t n_p \delta_{sq} \right), \tag{B3d}$$

$$(\hat{\xi}_3)_{stpqkm} = \frac{b}{2} \sum \frac{2n_t n_p n_q n_k \delta_{sm} - 5n_s n_t n_p n_q n_k n_m}{R}, \tag{B3e}$$

$$(\hat{\gamma}_{M1})_{stp} = b\varepsilon_{spm} \sum n_m n_t \left(\frac{1}{R} + \frac{k^2 R}{2} \right), \tag{B3f}$$

$$(\hat{\xi}_{M0})_{stp} = 3b^3 \varepsilon_{smp} \sum n_m n_t \left(\frac{1}{R^3} + \frac{k^2}{6R^2} \right), \tag{B3g}$$

$$(\hat{\xi}_{M2})_{stpqk} = \frac{3}{2}\, b\varepsilon_{smk} \sum \frac{n_m n_t n_p n_q}{R}. \tag{B3h}$$

Here $n_s \equiv (\vec{n})_s$, ε_{smp} is an antisymmetric unit tensor of the third rank and N_σ is the total number of particles inside the LS. In formulas (B3f), (B3g), (B3h) summation over the index m is implied.

For a cubic lattice in a coordinate system of crystallographic axes we have the tensors

$$(\hat{\gamma}_0)_{st} = \frac{2}{3}\left[k^2 b^3 \sum \frac{1}{R} + i(kb)^3 (N_\sigma - 1) \right] \delta_{st}, \tag{B4a}$$

$$(\hat{\gamma}_2)_{stpq}=\frac{b}{2}\sum\;\frac{(1-3n_x{}^4)\,\delta_{st}\delta_{pq}+\frac{1}{2}\left(\frac{1}{3}-3n_x^4\right)\delta_{sq}\delta_{ip}+\frac{3}{2}\,(5n_x{}^4-1)\,\delta_{st}\delta_{sp}\delta_{sq}}{R}, \tag{B4b}$$

$$(\hat{\xi}_1)_{stpq}=b^3\sum\left[\left(3\,\frac{5n_x{}^4-1}{R^3}+k^2\,\frac{3n_x{}^4-1}{2R}\right)\delta_{sq}\delta_{tp}+\frac{3}{2}\left(\frac{5}{R^3}+\frac{k^2}{12R}\right)(5n_x^4-1)\,\delta_{st}\delta_{sp}\delta_{sq}\right], \tag{B4c}$$

$$(\hat{\xi}_3)_{stpqkm}=\frac{b}{2}\sum\;\frac{1}{R}\left[5(-1+9n_x{}^4-6n_x{}^6)\,\delta_{st}\delta_{pk}\delta_{qm}+2(-2+21n_x{}^4-15n_x{}^6)\,\delta_{sk}\delta_{pq}\delta_{tm}+15\left(\frac{1}{2}-5n_x{}^4+\frac{7}{2}\,n_x{}^6\right)\right.$$
$$\times(\delta_{st}\delta_{pq}\delta_{pk}\delta_{pm}+2\,\delta_{st}\delta_{sp}\delta_{sk}\delta_{qm}+\delta_{st}\delta_{sk}\delta_{sm}\delta_{pq})+(4-45n_x{}^4+35n_x{}^6)\,\delta_{sk}\delta_{tp}\delta_{tq}\delta_{tm}+\left(-5+\frac{105}{2}\,n_x{}^4\right.$$
$$\left.\left.-\frac{77}{2}\,n_x{}^6\right)\delta_{st}\delta_{sp}\delta_{sq}\delta_{sk}\delta_{sm}\right], \tag{B4d}$$

$$(\hat{\gamma}_{M1})_{stp}=\frac{b}{3}\,\varepsilon_{stp}\sum\;\frac{1}{R}, \tag{B4e}$$

$$(\hat{\xi}_{M2})_{stpqk}=\frac{b}{2}\sum\;\frac{1}{R}\left[(1-3n_x{}^4)\varepsilon_{spk}\delta_{tq}+\frac{3}{2}\,(5n_x{}^4-1)\varepsilon_{spk}\delta_{tq}\delta_{tp}\right]. \tag{B4f}$$

In this case tensor $\hat{\xi}_{M0}$ is proportional to an antisymmetrical tensor $\hat{\varepsilon}$ so that its contraction with a symmetric quadrupole moment tensor is equal zero.

For a random medium an ensemble averaging allows one to replace summation in Eqs. (B3) or (B4) by integration so that

$$(\hat{\gamma}_0)_{st}=\frac{4\pi}{3}\left[k^2(a^2-b^2)+\frac{2i}{3}\,k^3(a^3-b^3)\right]\delta_{st}\,, \tag{B5a}$$

$$b^2(\hat{\gamma}_2)_{stpq}=\frac{2\pi}{5}\,(a^2-b^2)\left(\delta_{st}\delta_{pq}-\frac{1}{3}\,\delta_{sq}\delta_{tp}\right), \tag{B5b}$$

$$(\hat{\xi}_1)_{stpq}=\frac{2\pi}{5}\,k^2(b^2-a^2)\,\delta_{sq}\delta_{tp}\,, \tag{B5c}$$

$$b^2(\hat{\xi}_3)_{stpqkm}=\frac{2\pi}{7}\,(a^2-b^2)\left(\frac{2}{5}\,\delta_{sm}\delta_{tp}\delta_{qk}-\delta_{st}\delta_{pk}\delta_{qm}\right), \tag{B5d}$$

$$b^2(\hat{\gamma}_{M1})_{stp}=\frac{2\pi}{3}\,(a^2-b^2)\varepsilon_{stp}\,, \tag{B5e}$$

$$b^2(\hat{\xi}_{M2})_{stpqk}=\frac{2\pi}{5}\,k^2(a^2-b^2)\varepsilon_{spk}\delta_{tq}\,. \tag{B5f}$$

In the case of a jellylike medium the splitting procedure leads to the twofold decrease of the ''vacuum space'' sizes around the lth radiator, so that Eqs. (B5) remain true after the renormalization $b\rightarrow b/2$. Note that, strictly sreaking, Eqs. (A4) do not hold true anymore for near of the LS center radiators, since for them parameter b/R_{il} is not small. However, there is no sense to improve an accuracy of calculations in Eq. (A3) since in the vicinity of the LS center the angular distribution function is determined by the near-distant order and so depends on the concrete accepted model of the medium (for a discussion of this problem, see Ref. [26]).

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

In a manner similar to that addopted in I, Appendix B, and keeping $(ka)^3$ terms, one can deduce the useful relations

$$\nabla\times\int_\sigma^\Sigma \vec{F}Gd^3\vec{r}\,'=\int_\sigma^\Sigma \nabla\times\vec{F}Gd^3\vec{r}\,'-\frac{4\pi}{3}\,a^2(1+ika)\nabla\times\vec{F}, \tag{C1}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\times\vec{F}Gd^3\vec{r}=\int_\sigma^\Sigma \nabla\times\nabla\times\vec{F}Gd^3\vec{r}+\frac{8\pi}{3}\left[1+\frac{(ka)^2}{2}+\frac{i(ka)^3}{3}\right]\vec{F}+\frac{4\pi}{15}\,a^2(\Delta\vec{F}-\nabla\times\nabla\vec{F}), \tag{C2}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\times\nabla\times\vec{F}Gd^3\vec{r}=\int_\sigma^\Sigma \nabla\times\nabla\times\nabla\times\vec{F}Gd^3\vec{r}+\frac{4\pi}{3}\left[1-\frac{(ka)^2}{2}-\frac{2}{3}\,i(ka)^3\right]\nabla\times\vec{F}+\frac{2\pi}{15}\,a^2\Delta\nabla\times\vec{F}, \tag{C3}$$

$$\nabla\times\int_\sigma^\Sigma \nabla\cdot\hat{F}d^3\vec{r}=\int_\sigma^\Sigma \nabla\times\nabla\cdot\hat{F}Gd^3\vec{r}-\frac{4\pi}{3}\left[1+\frac{(ka)^2}{2}+\frac{i(ka)^3}{3}\right]\hat{\varepsilon}:\hat{F}-\frac{2\pi}{15}\,a^2[\Delta(\hat{\varepsilon}:\hat{F})+2\nabla\times\nabla\cdot\hat{F}], \tag{C4}$$

$$\begin{aligned}\nabla\times\int_\sigma^\Sigma \nabla\times\nabla\cdot\hat{F}Gd^3\vec{r}&=\int_\sigma^\Sigma \nabla\times\nabla\times\nabla\cdot\hat{F}G^3\vec{r}+\frac{4\pi}{15}\left\{\left[1+\frac{(ka)^2}{2}+\frac{i(ka)^3}{3}\right][\nabla\cdot(7\hat{F}+2\hat{F}^*)-3\nabla\mathrm{Tr}\hat{F}],\right.\\&\left.+(ka)^2\left[\nabla\cdot\left(2\hat{F}^*-\frac{\hat{F}}{2}\right)-\frac{1}{2}\,\nabla\mathrm{Tr}\hat{F}\right]\right\}+\frac{2\pi}{105}\,a^2[\Delta\nabla\cdot(11\hat{F}+4\hat{F}^*)\\&-3\Delta\nabla\mathrm{Tr}\hat{F}-6\nabla(\nabla\cdot\nabla\cdot\hat{F})],\end{aligned} \tag{C5}$$

$$\begin{aligned}\nabla\times\int_\sigma^\Sigma &\nabla\times\nabla\times\nabla\cdot\hat{F}Gd^3\vec{r}\\&=\int_\sigma^\Sigma \nabla\times\nabla\times\nabla\times\nabla\cdot\hat{F}Gd^3\vec{r}+\frac{4\pi}{5}\,\nabla\times\nabla\cdot(\hat{F}+\hat{F}^*)\\&+\frac{2\pi}{15}\,k^2\left\{-10\left[1+\frac{(ka)^2}{2}+i\,\frac{(ka)^3}{3}\right]\hat{\varepsilon}:\hat{F}+a^2[\nabla\right.\\&\left.\times\nabla\cdot(\hat{F}^*-\hat{F})-\Delta\hat{\varepsilon}:\hat{F}]\right\}+\frac{2\pi}{35}\,a^2\Delta\nabla\times\nabla\cdot(\hat{F}+\hat{F}^*).\end{aligned} \tag{C6}$$

Using the above equations one may perform the factoring of the operator $\nabla\times\nabla\times$ out of the integral sign, in Eqs. (5) and (30).

APPENDIX D: CALCULATION OF THE INTEGRALS OF $\vec{E}$ AND $\vec{H}$ OVER THE LS SURFACE

One can calculate the integral over the surface of the LS taking into account the terms $b^2\nabla\nabla\vec{E}$:

$$\begin{aligned}\int_\sigma\left(\vec{E}\,\frac{\partial G}{\partial\nu}-G\,\frac{\partial\vec{E}}{\partial\nu}-\vec{n}G\nabla'\cdot\vec{E}\right)d^2\vec{r}_\sigma&=4\pi e^{ika}\left[(1-ika)\vec{E}\right.\\&\left.+\frac{a^2}{3}\left(\frac{1-ika}{2}\,\Delta\vec{E}-\nabla\times\nabla\times\vec{E}\right)\right],\end{aligned} \tag{D1}$$

and a similar equation for $\vec{H}$. Then, by use of Eqs. (34) and (45) for macroscopic fields we may express their spatial derivatives through these fields and densities $\vec{P}$, $\hat{Q}$, and $\vec{M}$. By applying the operator $\nabla\times\nabla\times$ to both parts of Eq. (D1), we find

$$\begin{aligned}\frac{1}{4\pi k^2}\,\nabla\times\nabla\times\int_\sigma&\left(\vec{E}\,\frac{\partial G}{\partial\nu}-G\,\frac{\partial\vec{E}}{\partial\nu}-\vec{n}G\nabla'\cdot\vec{E}\right)d^2r_\sigma\\&=e^{ika}\left\{\left[1-ika-\frac{(ka)^2}{2}+i\,\frac{(ka)^3}{6}\right]\right.\\&\times\left[\vec{E}+4\pi\left(\vec{p}-\nabla\cdot\hat{Q}+\frac{i}{k}\,\nabla\times\vec{M}\right)\right]\\&\left.-2\pi\left(1-\frac{ika}{3}\right)a^2\nabla\times\nabla\times\left(\vec{P}-\nabla\cdot\hat{Q}+\frac{i}{k}\,\nabla\times\vec{M}\right)\right\},\end{aligned} \tag{D2}$$

and a similar expression for $\vec{H}$. Keeping an accuracy up to $(ka)^3$ we come to the relations

$$\begin{aligned}\nabla\times\nabla\times\int_\sigma&\left[\frac{1}{4\pi k^2}\left(\vec{E}\,\frac{\partial G}{\partial\nu}-G\,\frac{\partial\vec{E}}{\partial\nu}-\vec{n}G\nabla'\cdot\vec{E}\right)\right.\\&\left.+G\left(\frac{i}{k}\,\nabla\times\vec{M}-\hat{Q}\cdot\vec{n}\right)\right]d^2\vec{r}_\sigma\\&=\vec{E}+4\pi\left(\vec{P}-\nabla\cdot\hat{Q}+\frac{i}{k}\,\nabla\times\vec{M}\right)-2\pi a^2\nabla\times\nabla\\&\times\left[\left(1+\frac{2i}{3}\,ka\vec{P}-\frac{1}{3}\,\nabla\cdot\hat{Q}+\frac{i}{3k}\,\nabla\times\vec{M}\right],\end{aligned} \tag{D3}$$

$$\nabla\times\nabla\times\int_\sigma\Bigg[\frac{1}{4\pi k^2}\left(\vec{H}\,\frac{\partial G}{\partial\nu}-G\,\frac{\partial\vec{H}}{\partial\nu}-\vec{n}G\nabla'\cdot\vec{H}\right)+\frac{i}{k}\,([\vec{n}\cdot\hat{Q}\times\nabla' G]+G[\vec{n}\times\nabla'\cdot\hat{Q}]+G[\vec{P}\times\vec{n}])\Bigg]d^2\vec{r}_\sigma$$
$$=\vec{H}+4\pi\left(\vec{M}+\frac{i}{k}\,\nabla\times\nabla\cdot\hat{Q}-\frac{i}{k}\,\nabla\times\vec{P}\right)-2\pi a^2\times\nabla\times\left[\left(1+\frac{2i}{3}\,ka\right)\vec{M}+\frac{i}{k}\left(\frac{1}{5}\,\nabla\cdot\hat{Q}-\frac{1}{3}\,\nabla\times\vec{P}\right)\right]. \quad \text{(D4)}$$

APPENDIX E: CUBIC LORENTZ CAVITY

To simplify the calculations of the sums g_0, g_1, and g_2 for a cubic lattice medium it is convenient to change the form of the Lorentz cavity, i.e., to be exact, pass to the cavity of a cubic form. As is evident from Appendixes A and B, for finding the difference between the macroscopic and local fields it is necessary to calculate three volume integrals of the type $\int(n_x^m/R)d^3\vec{r}\,'$, where $m=0,4,6$, and three surface integrals $\int(\partial/\partial\nu)(n_x^m/R)d^2\vec{r}_\sigma$, were $\partial/\partial\nu$ is the derivative in the direction of the inward normal. For such a calculation let us start from the equality

$$\frac{\partial}{\partial x'}\,\frac{X^{m-1}}{R^{l+m-2}}=-(m-1)\,\frac{X^{m-2}}{R^{l+m-2}}+(l+m-2)\,\frac{X^m}{R^{l+m}},\qquad X=x-x' \quad \text{(E1)}$$

and then integrate both parts of Eq. (E1) over $\vec{r}\,'$. After that turn the volume integral on the left-hand part into the surface integral

$$\int_0^\sigma\frac{\partial}{\partial x'}\,\frac{X^{m-1}}{R^{l+m-2}}\,d^3\vec{r}\,'=\int_\sigma\frac{X^{m-1}}{R^{l+m-2}}\,(\vec{n}_\sigma)_x d^2\vec{r}_\sigma, \quad \text{(E2)}$$

where $\vec{n}_\sigma$ is the unit vector of the inward normal to the boundary σ of the volume of integration. As a result, we find

$$\int_0^\sigma\frac{X^m}{R^{l+m}}\,d^3\vec{r}\,'=\frac{m-1}{l+m-2}\int_0^\sigma\frac{X^{m-2}}{R^{l+m-2}}\,d^3\vec{r}\,'+\frac{1}{l+m-2}\int_\sigma\frac{X^{m-1}}{R^{l+m-2}}\,(\vec{n}_\sigma)_x d^2\vec{r}_\sigma. \quad \text{(E3)}$$

By repetition of such a procedure for the first right-hand term of Eq. (E3) we obtain, finally, for any value of m a set of the surface integrals and one volume integral $\int_0^\sigma(d^3\vec{r}\,'/R^l)$. By applying the formula (E3) to the case $m=2$ and using the obvious equality

$$\int_0^\sigma\frac{X^2}{R^l}\,d^3\vec{r}\,'=\frac{1}{3}\int_0^\sigma\frac{X^2+Y^2+Z^2}{R^l}\,d^3\vec{r}\,'\equiv\frac{1}{3}\int_0^\sigma\frac{d^3\vec{r}\,'}{R^{l-2}}, \quad \text{(E4)}$$

we can turn the last volume integral into the surface integral

$$\left(\frac{l}{3}-1\right)\int_0^\sigma\frac{1}{R^l}\,d^3\vec{r}\,'=\int_\sigma\frac{X}{R^l}\,(\vec{n}_\sigma)_x d^2\vec{r}_\sigma. \quad \text{(E5)}$$

For the case of a cubic cavity, Eq. (E3) can be rewritten as

$$\int_{-a}^a\int_{-a}^a\int_{-a}^a\frac{X^m dX\,dY\,dZ}{(X^2+Y^2+Z^2)^{(l+m)/2}}=\frac{m-1}{l+m-2}\int_{-a}^a\int_{-a}^a\int_{-a}^a\frac{X^{m-2}dXdYdZ}{(X^2+Y^2+Z^2)^{(l+m-2)/2}}-\frac{2a^{m-1}}{l+m-2}\int_{-a}^a\int_{-a}^a\frac{dYdZ}{(a^2+Y^2+Z^2)^{(l+m-2)/2}}. \quad \text{(E6)}$$

Here $Y\equiv y-y'$, $Z\equiv z-z'$, and $2a$ is the size of the cavity. Formula (E5) turns into the relation

$$\left(\frac{1}{l}-\frac{1}{3}\right)\int_{-a}^a\int_{-a}^a\int_{-a}^a\frac{dXdYdZ}{(X^2+Y^2+Z^2)^{l/2}}=\frac{2a}{l}\int_{-a}^a\int_{-a}^a\frac{dYdZ}{(a^2+Y^2+Z^2)^{1/2}}. \quad \text{(E7)}$$

These surface integrals can be calculated directly by use of the polar coordinates in the YZ plane. As a result, we obtain the necessary expressions

$$\int_0^\sigma\frac{1}{R}\,d^3\vec{r}\,'=3a^2B_1=2[6\ln(\sqrt{2}+3)-\pi]a^2, \quad \text{(E8a)}$$

$$\int_0^\sigma\frac{{n_x}^4}{R}\,d^3\vec{r}\,'=a^2\left(B_1-\frac{2}{3}\,B_3\right)=2\left[2\ln(\sqrt{2}+3)-\frac{5}{9}\,\pi\right]a^2, \quad \text{(E8b)}$$

$$\int_0^\sigma\frac{{n_x}^6}{R}\,d^3\vec{r}\,'=a^2\left(B_1-\frac{2}{3}\,B_3-\frac{2}{5}\,B_5\right)=2\left[2\ln(\sqrt{2}+3)-\frac{1}{5}\left(3\pi+\frac{4}{3\sqrt{3}}\right)\right]a^2, \quad \text{(E8c)}$$

$$\int_\sigma\frac{\partial}{\partial\nu}\,\frac{1}{R}\,d^2\vec{r}_\sigma=6B_3=4\pi, \quad \text{(E9a)}$$

$$\int_\sigma\frac{\partial}{\partial\nu}\,\frac{{n_x}^4}{R}\,d^2\vec{r}_\sigma=2(5B_7-4B_5+10D_4)=\frac{1}{3}\left(\frac{518}{\sqrt{3}}-\frac{265}{3}\,\pi\right), \quad \text{(E9b)}$$

$$\int_\sigma \frac{\partial}{\partial \nu}\frac{n_x{}^6}{R}\, d^2\vec{r}_\sigma = 2(7B_9 - 6B_7 + 14D_6)$$

$$= \frac{1}{15}\left(\frac{19\,187}{3\sqrt{3}} - \frac{2293}{2}\,\pi\right), \quad \text{(E9c)}$$

where

$$B_m = 4\int_0^1\int_0^1 \frac{du\,d\nu}{(1+u^2+\nu^2)^{m/2}}$$

$$= \frac{2}{m-2}\left[\pi - 2\int_0^{\pi/3}\left(\frac{\cos\xi}{1+\cos\xi}\right)^{(m-3)/2} d\xi\right], \quad \text{(E10a)}$$

$$B_1 = 2\left[2\ln(\sqrt{2}+3) - \frac{\pi}{3}\right], \quad \text{(E10b)}$$

$$B_3 = \frac{2\pi}{3}, \quad \text{(E10c)}$$

$$B_5 = \frac{2}{3}\left(\frac{\pi}{3} + \frac{2}{\sqrt{3}}\right), \quad \text{(E10d)}$$

$$B_7 = \frac{2}{15}\left(\pi + \frac{26}{3\sqrt{3}}\right), \quad \text{(E10e)}$$

$$B_9 = \frac{2}{21}\left(\pi + \frac{148}{15\sqrt{3}}\right), \quad \text{(E10f)}$$

$$D_4 \equiv 4\int_0^1\int_0^1 \frac{u^4\,du\,d\nu}{(1+u^2+\nu^2)^{7/2}} = \frac{1}{10}\left(\frac{773}{9\sqrt{3}} - \frac{29}{2}\,\pi\right), \quad \text{(E11a)}$$

$$D_6 \equiv 4\int_0^1\int_0^1 \frac{u^6\,du\,d\nu}{(1+u^2+\nu^2)^{9/2}} = \frac{1}{84}\left(\frac{19\,219}{15\sqrt{3}} - \frac{457}{2}\,\pi\right). \quad \text{(E11b)}$$

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