Method of integral equations and an extinction theorem for two-dimensional problems in nonlinear optics

A. V. Ghiner* and G. I. Surdutovich†
Instituto de Física e Química de São Carlos, Universidade de São Paulo, Caixa Postal 369, 13560-970 São Carlos, São Paulo, Brazil
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An approach using the generalized method of integral equations by substitution of the variables in the integral equation is applied to two- and quasi-two-dimensional systems. As a result, the connection between the integral and Maxwell equations as well as an extinction theorem for this case are established. The technique developed may be applied to any composite medium with a columnlike mesostructure. By use of the elementary cylinder radiator ("mesoscopic atom") concept we reduce the problem of finding the optical properties of such media to the calculation of the susceptibility of a dense two-dimensional gas. The calculated optical anisotropy depends dramatically not only on the concentration but also on the form of the inclusions (mesostructure). Our calculations of the dielectric permittivity tensor for a two-dimensional composite medium with wire mesostructure show excellent agreement with the experimental measurements of the long-wavelength dielectric constants for two orthogonal polarizations in a photonic crystal made of dielectric rods [W. M. Robertson *et al.*, J. Opt. Soc. Am. B **10**, 322 (1993)].

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I. INTRODUCTION

First we qualify the term two-dimensional (2D) system. Often this term is used for the so-called monolayer or very thin film with thickness negligibly small compared to the wavelength. Here we apply this term to another limiting case of optically very thick homogeneous films consisting of long threads (wires) oriented perpendicularly to the film's surface. As is known, in certain cases 2D systems have critically different properties in comparison with three-dimensional media. Therefore, it seems challenging to establish an extinction theorem and the relation between macro- and microcharacteristics of the medium starting from the fundamental principles of optics.

From a practical point of view one may cite at least three interesting physical 2D systems. First we mention Langmuir-Blodgett (LB) films and those self-assembled from long oriented molecules [1]. When the film thickness is larger than the wavelength and its physical characteristics do not depend on the z coordinate perpendicular to the surface (or when propagation of light has a waveguided character), then such a structure may be considered as two dimensional. The second example of quasi-two-dimensional structures is composite media, where one fraction is distributed inside the other in the form of parallel or quasiparallel columns. It is generally accepted that such a situation occurs, for example, in porous silicon—a new promising microelectronic material formed by electrochemical etching of crystalline Si. Its structure consists of undulating columns of crystalline Si with large diameters in comparison to interatomic distances. In the case of small porosity the structure is better described by vacuum columns in bulk crystalline Si [2]. The third example is related to a lattice composed of dielectric "atoms" [photonic band-gap (PBG) structure] [3]. Two-dimensional PBG structures consisting of arrays of dielectric rods in air have been proposed and studied [4]. In this case correct consideration of the local-field effects seems to be essential.

To solve the above-mentioned problems we applied here the method of integral equations (MIEs) [5], which allows a consecutive description of the process of light propagation in a medium with discrete structure. This method reveals a connection between linear and nonlinear micro- and macrocharacteristics of the optical media with an arbitrary structure when not only dipole but electric quadrupole and magnetic dipole moments of the elementary radiators are taken into account [6]. In the optical region the contribution of the last two mechanisms to the linear susceptibility is, as a rule, small in comparison with the dipole contribution, but they may be essential for nonlinear effects.

In Sec. II we put forward the basis idea for applying the MIEs to a calculation of the optical characteristics of composite media. It is a concept of a "mesoscopic radiator" in which, instead of a straightforward solution of the Maxwell equations for an inhomogeneous composite medium, we consider spherical or cylindrical inclusions of one medium in the other as the elementary radiators [7]. Due to the fact that dielectric cylinders (spheres) in an external homogeneous field behave like ideal two- (three-) dimensional dipoles, the MIEs may be applied to

*Permanent address: Automation and Electrometry Institute, 630090, Novosibirsk, Russia.
†Permanent address: Semiconductor Physics Institute, 630090, Novosibirsk, Russia.

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this situation. Such an approach corresponds to the notion of mesoscopic quantities averaged over the interatomic, but not over the intercylinder (sphere), distances [8]. We note that in reality the considered problems are not quite two dimensional since, although all the physical quantities are implied to be independent of the z coordinate oriented along the cylinders' axes, we will be interested in the determination of the electromagnetic field and z component of the electric, magnetic, and quadrupolar moments as well.

In Sec. III we apply the earlier [6,7] developed method of substitution of the variables in the integral equation, which in this case corresponds to the passage from the local (mesoscopic) field to the macroscopic one. This method proved to be effective for two-dimensional problems as well. We show the connection between the integral and Maxwell equations approaches and deduce an extinction theorem. The local-field problem for isotropic and anisotropic two-dimensional media is discussed.

These general results are applied in Sec. IV for two-dimensional composite media and self-assembled films. Finally, in Sec. V we summarize our results.

II. A CONCEPTION OF "TWO-DIMENSIONAL" ELEMENTARY RADIATORS (ELECTRIC DIPOLE, ELECTRIC QUADRUPOLE, AND MAGNETIC DIPOLE) AND THE FIELDS FROM THESE RADIATORS

By use of the Green's-function method for the wave equation in a vacuum we may write the Fourier transforms at frequency ω of the electric $\mathbf{E}$ and magnetic $\mathbf{H}$ field components of the two-dimensional dipole $\mathbf{d}$, quadrupole $\hat{s}$, and magnetic dipole $\mathbf{w}$ densities in the form analogous to the three-dimensional case [6]:

$$\mathbf{E}_d(\mathbf{r})=\nabla\times\nabla\times\mathbf{d}(\mathbf{r}')G(kR)\ , \tag{1a}$$

$$\mathbf{H}_d=-ik\nabla\times\mathbf{d}(\mathbf{r}')G(kR)\ , \tag{1b}$$

$$\mathbf{E}_s(\mathbf{r})=-\nabla\times\nabla\times\nabla\cdot\hat{s}(\mathbf{r}')G(kR)\ , \tag{2a}$$

$$\mathbf{H}_s=ik\nabla\times\nabla\cdot\hat{s}(\mathbf{r}')G(kR)\ , \tag{2b}$$

$$\mathbf{E}_w(\mathbf{r})=ik\nabla\times\mathbf{w}(\mathbf{r}')G(kR)\ , \tag{3a}$$

$$\mathbf{H}_w=\nabla\times\nabla\times\mathbf{w}(\mathbf{r}')G(kR)\ , \tag{3b}$$

where $\mathbf{R}=\mathbf{r}-\mathbf{r}'$ and $k=\omega/c$. Here two-dimensional (in the xy plane) vectors $\mathbf{r}$ and $\mathbf{r}'$ correspond to the coordinates of the observation point and of the radiator, respectively. $G(kR)=\pi i H_0^{(1)}(kR)$ is the Green's function of the scalar wave equation [9] and $H_0^{(1)}$ is the Hankel function of the first kind, zeroth order. The gradient symbol ∇ indicates differentiation over $\mathbf{r}$. A centered dot indicates the contraction of two tensors over the pair of indices.

As in the three-dimensional case [6], it follows from (2) that we always can add to tensor $\hat{s}$ a unit tensor multiplied by an arbitrary scalar function without changing the fields. This means we can choose a tensor $\hat{s}$ such that $\mathrm{Tr}\hat{s}=0$. Then from (2) and (3) it follows that $\hat{s}$ is a symmetric tensor, since the antisymmetric part can be removed by means of renormalization of the magnetic moment.

Performing the differentiation in (1) and assuming that $\partial/\partial z=0$ for all physical quantities, we arrive at formulas for the fields where contributions from the different radiation zones are separated:

$$\mathbf{E}_d=i\pi k\frac{H_1^{(1)}}{R}[2\mathbf{n}(\mathbf{n}\cdot\mathbf{d})-\mathbf{d}]+i\pi k^2H_0^{(1)}[\mathbf{d}-\mathbf{n}(\mathbf{n}\cdot\mathbf{d})]\ , \tag{4a}$$

$$E_{dz}=i\pi k^2H_0^{(1)}d_z\ , \tag{4b}$$

$$\mathbf{H}_d=-i\pi k^2H_1^{(1)}\hat{\sigma}_y\cdot\mathbf{n}d_z\ , \tag{4c}$$

$$H_{dz}=-\pi k^2H_1^{(1)}[\mathbf{n}\times\mathbf{d}]_z\ , \tag{4d}$$

$$\mathbf{E}_s=2\frac{i\pi k}{R}\left[2\frac{H_1^{(1)}}{R}-kH_0^{(1)}\right][2\mathbf{n}(\mathbf{n}\cdot\mathbf{n}\cdot\hat{s})-\mathbf{n}\cdot\hat{s}]$$
$$+i\pi k^3H_1^{(1)}[\mathbf{n}\cdot\hat{s}-\mathbf{n}(\mathbf{n}\cdot\mathbf{n}\cdot\hat{s})]\ , \tag{5a}$$

$$E_{sz}=i\pi k^3H_1^{(1)}(\mathbf{n}\cdot\hat{s})_z\ , \tag{5b}$$

$$\mathbf{H}_s=-\pi k^2\left[2\frac{H_1^{(1)}}{R}-kH_0^{(1)}\right]\mathbf{n}\times(\mathbf{n}\cdot\hat{s})+i\pi k^2\frac{H_1^{(1)}}{R}\hat{\sigma}_y\cdot\mathbf{s}_z\ , \tag{5c}$$

$$H_{sz}=-\pi k^2\left[2\frac{H_1^{(1)}}{R}-kH_0^{(1)}\right][\mathbf{n}\times(\mathbf{n}\cdot\hat{s})]_z\ , \tag{5d}$$

$$\mathbf{E}_w=i\pi k^2H_1^{(1)}\hat{\sigma}_y\cdot\mathbf{n}w_z\ , \tag{6a}$$

$$E_{wz}=\pi k^2H_1^{(1)}[\mathbf{n}\times\mathbf{w}]_z\ , \tag{6b}$$

$$\mathbf{H}_w=i\pi k\frac{H_1^{(1)}}{R}[2\mathbf{n}(\mathbf{n}\cdot\mathbf{w})-\mathbf{w}]+i\pi k^2H_0^{(1)}[\mathbf{w}-\mathbf{n}(\mathbf{n}\cdot\mathbf{w})]\ , \tag{6c}$$

$$H_{wz}=i\pi k^2H_0^{(1)}w_z\ , \tag{6d}$$

where $\mathbf{n}\equiv\mathbf{R}/R$ and $\mathbf{s}_z\equiv(s_{xz},s_{yz})$. Here $\mathbf{n}$ stands for a two-dimensional vector with components n_x,n_y. In the contraction operation of the unit vector $\mathbf{n}$ with tensor $\hat{s}$ this vector is considered at first as a three-dimensional vector with component $n_z=0$ and then, after this contraction, the z component of the three-dimensional vector obtained is omitted. The operator $\hat{\sigma}_y$ indicates the Pauli y matrix

$$\hat{\sigma}_y=\begin{bmatrix}0 & -i\\ i & 0\end{bmatrix} \tag{7}$$

and $H_1^{(1)}$ is the first-order Hankel function of the first kind. Since zx, zy, and zz components of the quadrupole tensor do not enter into the expressions for fields then without any limitation of generality we may set $s_{zz}=0$, and then not only $\mathrm{Tr}\hat{s}=0$ but the equality $s_{xx}+s_{yy}=0$ is satisfied as well.

Just like in the three-dimensional case we have now the separation of contributions from various terms into various zones. For example, in Eq. (4a) for dipole radiation the first term describes near-distant (static) and the intermediate zones whereas the second term describes the wave (radiation) zone. It is noteworthy that the z component of the electric field does not have a static term. It is connected with the obvious fact that the static field is perpendicular to the axis z of a charge thread. This assertion is also true for other z components, namely, the quadrupole's electric field and the magnetic dipole's magnetic field. Finally, in the case of a composite medium, densities $\mathbf{d}$, $\hat{s}$, and $\mathbf{w}$ are replaced by integrals over the cylinder's cross section.

III. THE LOCAL-FIELD PROBLEM AND AN EXTINCTION THEOREM UNDER PROPAGATION OF LIGHT IN TWO-DIMENSIONAL OPTICAL MEDIA

Equations (4)–(6) allow us to calculate the electric and magnetic fields acting on the radiator at point $\mathbf{r}$ due to all other radiators and the incident wave. By introducing the "Lorentz cavity"—in our case it is a cylinder σ of radius a with axis parallel to the z axis—and choosing the parameters a and b in the following manner:

$$\lambda \gg a \gg b \ , \tag{8}$$

where b is the characteristic distance between the radiators (cylinders) and λ is the wavelength; for radiators inside the Lorentz cavity we may pass from a summation to the integration by analogy with the three-dimensional case. As a result, we come to the following integral equation for the electric field $\mathbf{E}'$ and the magnetic field $\mathbf{H}'$ acting on the elementary radiator:

$$\mathbf{E}'(\mathbf{r}')=\mathbf{E}_i(\mathbf{r})+\mathbf{E}_\sigma(\mathbf{r}') + \int_\sigma^\Sigma (\nabla\times\nabla\times\mathbf{P}G-\nabla\times\nabla\times\nabla\cdot\hat{Q}G + ik\nabla\times\mathbf{M}G)d^2\mathbf{r}' \ , \tag{9a}$$

$$\mathbf{H}'(\mathbf{r}')=\mathbf{H}_i(\mathbf{r})+\mathbf{H}_\sigma(\mathbf{r}') + \int_\sigma^\Sigma (-ik\nabla\times\mathbf{P}G+ik\nabla\times\nabla\cdot\hat{Q}G + ik\nabla\times\nabla\times\mathbf{M}G)d^2\mathbf{r}' \ , \tag{9b}$$

where $\mathbf{P}$, $\mathbf{M}$, and $\hat{Q}$ are the electric dipole, magnetic dipole, and quadrupole volume densities, respectively; $\mathbf{E}_i$ and $\mathbf{H}_i$ are the strengths of the electric and magnetic fields of the incident wave, σ is the Lorentz cylinders' surface, and Σ is the boundary of the medium.

Here $\mathbf{E}_\sigma$ and $\mathbf{H}_\sigma$ are the contributions from the radiators inside the Lorentz cavity. It is necessary to emphasize that, while calculating the terms $\mathbf{E}_\sigma$ and $\mathbf{H}_\sigma$, we cannot pass from summation to integration because for the radiators disposed not far from the observation point $\mathbf{r}$, discreteness of the medium is important. At the same time, the changes of $\mathbf{P}$, $\hat{Q}$, and $\mathbf{M}$ inside the Lorentz cavity may be considered to be small due to condition (8). Then expressions for $\mathbf{E}_\sigma$ and $\mathbf{H}_\sigma$ take the form [compare them with formulas (7) and (8) of Ref. [6] for the three-dimensional case]

$$\mathbf{E}_\sigma(\mathbf{r})=\hat{\gamma}\cdot\mathbf{P}+\frac{1}{b}\hat{\xi}:\hat{Q}+ikb\hat{\gamma}_M\cdot\mathbf{M}+b\hat{\gamma}_1:(\nabla\mathbf{P})+\hat{\xi}_1\vdots(\nabla\hat{Q}) \ , \tag{10a}$$

$$\mathbf{H}_\sigma(\mathbf{r})=\hat{\gamma}\cdot\mathbf{M}+ik\hat{\xi}_M:\hat{Q}-ikb\hat{\gamma}_M\cdot\mathbf{P}+b\hat{\gamma}_1:(\nabla\mathbf{M}) \ . \tag{10b}$$

With allowance of Eqs. (4)–(8) for tensors $\hat{\gamma}$ and $\hat{\xi}$, we obtain the following expressions:

$$(\hat{\gamma})_{st}=2b^2\sum_{j\neq l}^{\sigma}\frac{2(\mathbf{n}_{lj})_s(\mathbf{n}_{lj})_t-(1-\delta_{sz})\delta_{st}}{R_{lj}^2} \ , \tag{11a}$$

$$(\hat{\gamma}_1)_{stp}=-2b\sum_{j\neq l}^{\sigma}\frac{2(\mathbf{n}_{lj})_s(\mathbf{n}_{lj})_t(\mathbf{n}_{lj})_p-(1-\delta_{sz})\delta_{sp}(\mathbf{n}_{lj})_t}{R_{lj}} \ , \tag{11b}$$

$$(\hat{\xi})_{stp}=8b^3\sum_{j\neq l}^{\sigma}\frac{2(\mathbf{n}_{lj})_s(\mathbf{n}_{lj})_t(\mathbf{n}_{lj})_p-(1-\delta_{sz})\delta_{sp}(\mathbf{n}_{lj})_t}{R_{lj}^3} \ , \tag{11c}$$

$$(\hat{\xi}_1)_{stpq}=-8b^2\sum_{j\neq l}^{\sigma}\frac{2(\mathbf{n}_{lj})_s(\mathbf{n}_{lj})_t(\mathbf{n}_{lj})_p(\mathbf{n}_{lj})_q-(1-\delta_{sz})\delta_{sq}(\mathbf{n}_{lj})_t(\mathbf{n}_{lj})_p}{R_{lj}^2} \ , \tag{11d}$$

$$(\hat{\gamma}_M)_{st}=-2ikb^2(\hat{\epsilon})_{sqt}\sum_{j\neq l}^{\sigma}\frac{(\mathbf{n}_{lj})_q}{R_{lj}} \ , \tag{11e}$$

$$(\hat{\xi}_M)_{stp}=2b^2\sum_{j\neq l}^{\sigma}\frac{2(\hat{\epsilon})_{sqp}(\mathbf{n}_{lj})_q(\mathbf{n}_{lj})_t-(\hat{\epsilon})_{stp}\delta_{pz}}{R_{lj}^2} \ . \tag{11f}$$

Here $\mathbf{n}_{lj} \equiv \mathbf{R}_{lj}/R_{lj}$ and $\mathbf{R}_{lj} \equiv \mathbf{r}_l - \mathbf{r}_j$. The indices s,t,p number the Cartesian components, δ_{st} is the Kronecker symbol, and $\hat{\epsilon}$ is the antisymmetric unit third-rank tensor. In Eqs. (11e) and (11f) summation over the index q is implied. The radius vectors $\mathbf{r}_l$ and $\mathbf{r}_j$ determine the position of the Lorentz cavity's center and location of the radiator in the xy plane [components $(\mathbf{r}_l)_z$ and $(\mathbf{r}_j)_z$ are always considered to be zero]. Note that, as in the three-dimensional case, Eqs. (10) and (11) hold true with an accuracy better than the electrostatic field approximation, being correct up to terms of first order in the parameter kR inclusive.

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

$$\hat{\gamma} = \hat{\gamma}_1 = \hat{\gamma}_M = \hat{\zeta} = \hat{\zeta}_1 = \hat{\zeta}_M = 0 \; . \tag{12}$$

Vanishing of tensors $\hat{\gamma}$ and $\hat{\zeta}$ under random distribution of the radiators removes the obstacle of divergence at the upper limit of the summation in Eq. (11).

To calculate the macroscopic parameters we perform, according to [6,7], a substitution of the variables in the integral equation. As was shown earlier, this substitution corresponds to the passage from local (in our case mesoscopic) fields $\mathbf{E}', \mathbf{H}'$ to the macroscopic fields $\mathbf{E}, \mathbf{H}$ by the formulas

$$\begin{aligned} \mathbf{E} &= \mathbf{E}' + \hat{\beta}\cdot\mathbf{P} + \frac{1}{b}\hat{\eta}:\hat{Q} + ik\hat{\beta}_M\cdot\mathbf{M} + b\hat{\beta}_1:(\nabla\mathbf{P}) \\ &\quad + \hat{\eta}_1 \vdots (\nabla\hat{Q}) + \frac{i}{k}\hat{\beta}_{M1}:(\nabla\mathbf{M}) \; , \\ \mathbf{H} &= \mathbf{H}' + \hat{\beta}\cdot\mathbf{M} + ik\hat{\eta}_M:Q - ik\hat{\beta}_M\cdot\mathbf{P} + b\hat{\beta}_1:(\nabla\mathbf{M}) \\ &\quad + \frac{i}{k}\hat{\eta}_{M2}\vdots(\nabla\nabla\hat{Q}) - \frac{i}{k}\hat{\beta}_{M1}:(\nabla\mathbf{P}) \; , \end{aligned} \tag{13}$$

where $\hat{\beta}$ and $\hat{\eta}$ are free parameters, which we choose in such a manner that fields $\mathbf{E}$ and $\mathbf{H}$, together with the integral equations (9), satisfy also the following wave equations:

$$\begin{aligned} \nabla\times\nabla\times\mathbf{E} - k^2\mathbf{E} &= 4\pi g_E k^2 \left[\mathbf{P} - \nabla\cdot\hat{Q} + \frac{i}{k}\nabla\times\mathbf{M} \right] , \\ \nabla\times\nabla\times\mathbf{H} - k^2\mathbf{H} &= 4\pi g_M k^2 \left[\mathbf{M} + \frac{i}{k}\nabla\times\nabla\cdot\hat{Q} - \frac{i}{k}\nabla\times\mathbf{P} \right] , \end{aligned} \tag{14}$$

where g_E and g_M are free parameters.

Using the results of Appendix B we may, just as in Ref. [6], transform integral equations (9) to the following form:

$$\begin{aligned} \left[1 - \frac{1}{g_E}\right]\mathbf{E} &= (\hat{\gamma} + \hat{\beta} - \hat{\Psi} + 4\pi)\cdot\mathbf{P} + \frac{1}{b}(\hat{\eta} + \hat{\zeta}):\hat{Q} + ikb(\hat{\gamma}_M + \hat{\beta}_M)\cdot\mathbf{M} + b(\hat{\gamma}_1 + \hat{\beta}_1):(\nabla\mathbf{P}) \\ &\quad + (\hat{\zeta}_1 + \hat{\eta}_1 + \hat{\Theta}_{11} - 4\pi\hat{\delta}_4)\vdots(\nabla\hat{Q}) + \frac{i}{k}(\hat{\beta}_{M1} - \hat{\Psi}_{11} + 4\pi\hat{\epsilon}):(\nabla M) \\ &\quad + \mathbf{E}_i + \nabla\times\nabla\times\int_\Sigma \left[\frac{1}{4\pi k^2}\left[\mathbf{E}\frac{\partial G}{\partial\nu} - G\frac{\partial\mathbf{E}}{\partial\nu} + G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E} \right] + G\left[\hat{Q}\cdot\mathbf{n}_\Sigma + \frac{i}{k}[\mathbf{M}\times\mathbf{n}_\Sigma] \right] \right] d\mathbf{r}_\Sigma \; , \end{aligned} \tag{15a}$$

$$\begin{aligned} \left[1 - \frac{1}{g_H}\right]\mathbf{H} &= (\hat{\gamma} + \hat{\beta} - \hat{\Psi} + 4\pi)\cdot\mathbf{M} + ik(\hat{\zeta}_M + \hat{\eta}_M - \hat{\Theta}_2):\hat{Q} - ikb(\hat{\gamma}_M + \hat{\beta}_M)\cdot\mathbf{P} + b(\hat{\gamma}_1 + \hat{\beta}_1):(\nabla\mathbf{M}) \\ &\quad + \frac{i}{k}(\hat{\eta}_{M2} - \hat{\Theta}_{22} + 4\pi\hat{\delta}_5)\vdots(\nabla\nabla\hat{Q}) - \frac{i}{k}(\hat{\beta}_{M1} - \hat{\Psi}_{11} + 4\pi\hat{\epsilon}):(\nabla\mathbf{P}) \\ &\quad + \mathbf{H}_i + \nabla\times\nabla\times\int_\Sigma \left[\frac{1}{4\pi k^2}\left[\mathbf{H}\frac{\partial G}{\partial\nu} - G\frac{\partial\mathbf{H}}{\partial\nu} + G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{H} \right] - \frac{i}{k}(\mathbf{n}_\Sigma\cdot\hat{Q}\times\nabla' G + G\mathbf{n}_\Sigma\times\nabla'\cdot\hat{Q} + G\mathbf{P}\times\mathbf{n}_\Sigma) \right] d\mathbf{r}' , \end{aligned} \tag{15b}$$

where tensors $\hat{\Psi}$ and $\hat{\Theta}$ are due to terms arising after factoring the operator $\nabla\times\nabla\times$ from the integrand. The differentiation operation over $\mathbf{r}$ and the operation of integration over $\mathbf{r}'$ do not commute because the lower limit of integration depends on $\mathbf{r}$. Here $\hat{\delta}_4$ is a symmetric fourth-rank unit tensor

$$(\hat{\delta}_4)_{ijkl} \equiv \delta_{il}\delta_{jk} \; , \quad \hat{\delta}\vdots(\nabla\hat{Q}) = \nabla\cdot\hat{Q} \tag{16}$$

and $\hat{\delta}_5$ is a unit tensor of fifth rank

$$(\hat{\delta}_5)_{ijklm} = \epsilon_{ijm}\delta_{kl} \; , \quad \hat{\delta}_5\vdots(\nabla\nabla\hat{Q}) = \nabla\times\nabla\cdot\hat{Q} \; . \tag{17}$$

If we choose the values of the free parameters in the following way:

$$\begin{aligned} & g_E = g_H = 1 \; , \quad \hat{\beta} = -4\pi + \hat{\Psi} - \hat{\gamma} \; , \quad \hat{\eta} = -\hat{\zeta} \; , \quad \hat{\beta}_1 = -\hat{\gamma}_1 \; , \\ & \hat{\beta}_M = -\hat{\gamma}_M \; , \quad \hat{\eta}_1 = 4\pi\hat{\delta}_4 - \hat{\Theta}_{11} - \hat{\zeta}_1 \; , \quad \hat{\eta}_M = \hat{\Theta}_2 - \hat{\zeta}_M \; , \\ & \hat{\beta}_{M1} = -4\pi\hat{\epsilon} + \hat{\Psi}_{11} \; , \quad \hat{\eta}_{M2} = \hat{\Theta}_{22} - 4\pi\hat{\delta}_5 \; , \end{aligned} \tag{18}$$

then it is evident that all the extra-integral terms in (15), except $\mathbf{E}_i$ and $\mathbf{H}_i$, vanish. As a consequence Eqs. (15) take the form

$$\mathbf{E}_i+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2}\left[\mathbf{E}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{E}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E}\right]+G\left[\hat{Q}\cdot\mathbf{n}_\Sigma+\frac{i}{k}\left[\mathbf{M}\times\mathbf{n}_\Sigma\right]\right]\right]d\mathbf{r}_\Sigma=0\ , \tag{19a}$$

$$\mathbf{H}_i+\nabla\times\nabla\times\int_\Sigma\left[\frac{1}{4\pi k^2}\left[\mathbf{H}\frac{\partial G}{\partial\nu}-G\frac{\partial\mathbf{E}}{\partial\nu}+G\mathbf{n}_\Sigma\nabla'\cdot\mathbf{E}\right]-\frac{i}{k}([\mathbf{n}_\Sigma\hat{Q}\times\nabla'G]+G[\mathbf{n}_\Sigma\times\nabla'\cdot\hat{Q}]+G[\mathbf{P}\times\mathbf{n}_\Sigma])\right]d\mathbf{r}_\Sigma=0\ , \tag{19b}$$

where $d\mathbf{r}_\Sigma$ is the differential along the boundary line in the xy plane.

This is an extinction theorem for optical media, which may be considered to be two dimensional. For media with a "blurred" (compared to b) boundary, the terms with $\hat{Q}$ and $\hat{M}$ in (19a) and with $\hat{Q}$ and $\hat{P}$ in (19b) may be omitted. The expressions for the fields outside the medium (reflected waves) coincide with the left-hand parts of Eqs. (19).

After substitution of expressions (18) into (13) and (9) we come to the conclusion that the quantities $\mathbf{E}$ and $\mathbf{H}$ satisfy the system of macroscopic Maxwell equations, where vector $\mathbf{D}$ is given by the relation

$$\mathbf{D}=\mathbf{E}+4\pi\mathbf{P}-4\pi\nabla\cdot\hat{Q}\ . \tag{20}$$

Finally, for a nonmagnetic medium, in the linear approximation when $\mathbf{M}$ does not depend on $\mathbf{E}'$, we may set

$$\mathbf{d}=\hat{\alpha}\mathbf{E}'+\mathbf{d}^{NL} \tag{21}$$

and using Eqs. (13), (14), and (18) obtain the macroscopic equation

$$\nabla\times\nabla\times\mathbf{E}-k^2\hat{\epsilon}\cdot\mathbf{E}=4\pi k^2[\hat{f}_p\cdot\mathbf{P}^{NL}+\hat{f}_Q\cdot(\nabla\cdot\hat{Q})+\frac{i}{k}\hat{f}_M\nabla\times\mathbf{M}-\hat{f}_pN\hat{\alpha}\mathbf{F}]\ , \tag{22}$$

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

$$\hat{\epsilon}=1+4\pi[1-N\hat{\alpha}(4\pi-\hat{\Psi}+\hat{\gamma})]^{-1}\cdot N\hat{\alpha}\ , \tag{23}$$

$$\hat{f}_p=[1-N\hat{\alpha}(4\pi-\hat{\Psi}+\hat{\gamma})]^{-1}\ ,\quad \hat{f}_Q=\hat{\delta}_4-f_p\cdot N\hat{\alpha}\cdot\hat{\eta}_1\ , \tag{24}$$

$$\hat{f}_M=\hat{\epsilon}-\hat{f}_p\cdot N\hat{\alpha}\cdot(4\pi\hat{\epsilon}-\hat{\Psi}_{11})\equiv\hat{\epsilon}\ , \tag{25}$$

$$\hat{F}=\frac{1}{b}\hat{\xi}:\hat{Q}+ik\hat{\gamma}_M\cdot\mathbf{M}+b\hat{\gamma}_1:(\nabla\mathbf{P})\ . \tag{26}$$

In Eq. (25) $\hat{\epsilon}$ is a unit antisymmetric third-rank tensor.

In the case of random distribution of cylinders in the xy plane the treatment of dipole density $\mathbf{P}$, taking into account only terms linear in $\mathbf{E}'$ and $\mathbf{H}'$ in the density $\hat{Q}$, leads to not so cumbersome formulas. Consideration of vector invariance suggests the following natural linear dependence of $\hat{Q}$ from $\mathbf{E}'$:

$$\hat{Q}^{(L)}=\frac{N}{k^2}\hat{\alpha}_Q:(\nabla\mathbf{E}')\ , \tag{27}$$

where $\hat{\alpha}_Q$ is the fourth-rank tensor symmetric over the first and last pairs of indices.

In the case of an isotropic medium in the xy plane and taking into account the condition $(\nabla\cdot\mathbf{E}')=0$ we may write tensor $\hat{\alpha}$ in the following form:

$$\alpha_{ijkl}=\alpha_{Qi}\delta_{il}\delta_{jk}\ , \tag{28}$$

where

$$\alpha_{Qx}=\alpha_{Qy}\equiv\alpha_Q\ . \tag{29}$$

In the case considered the electric dipole and magnetic dipole polarizations have analogous properties. As a result, we can write the microscopic material equations. For convenience we do not present the connection formulas between $\mathbf{d},\hat{s},\mathbf{w}$ and $\mathbf{E}',\mathbf{H}'$, but give identical formulas for the bulk quantities $\mathbf{P},\hat{Q},\mathbf{M}$ and $N\mathbf{E}',N\mathbf{H}'$:

$$\begin{aligned}&\mathbf{P}=N\hat{\alpha}\cdot\mathbf{E}'+\mathbf{P}^{NL}\ ,\\&\nabla\cdot\hat{Q}=-N\hat{\alpha}_Q\cdot\mathbf{E}'+\nabla Q^{NL}\ ,\\&\mathbf{M}=N\hat{\alpha}_M\cdot\mathbf{H}'+\mathbf{M}^{NL}\ ,\\&(\hat{\alpha})_{ij}=\alpha_i\delta_{ij}\ ,\quad \alpha_x=\alpha_y=\alpha\ .\end{aligned} \tag{30}$$

For α_M there are analogous expressions. Finally, the relations between the macroscopic $\mathbf{E},\mathbf{H}$ and local $\mathbf{E}',\mathbf{H}'$ fields take the form

$$\begin{aligned}&(\mathbf{E})_{x,y}=(\mathbf{E}'-2\pi\mathbf{P}+2\pi\nabla\cdot\hat{Q})_{x,y}\ ,\quad E_z=E_z'\ ,\quad H_z=H_z'\ ,\\&H_x=H_x'-2\pi M_x+2\pi\frac{i}{k}\frac{\partial}{\partial y}(\nabla\cdot\hat{Q})_z-2\pi ikQ_{yz}\ ,\\&H_y=H_y'-2\pi M_y-2\pi\frac{i}{k}\frac{\partial}{\partial x}(\nabla\cdot\hat{Q})_z+2\pi ikQ_{xz}\ .\end{aligned} \tag{31}$$

Through the use of (30) we may eliminate from these expressions the linear parts P^L, Q^L, and M^L. Limiting ourselves to the case $Q_{iz}=0$ we obtain

$$\begin{aligned}&(\mathbf{E}')_{x,y}=\left[\frac{\mathbf{E}+2\pi\mathbf{P}^{NL}-2\pi(\nabla\cdot\hat{Q}^{NL})}{1-2\pi N\alpha_p-2\pi N\alpha_Q}\right]_{x,y}\ ,\\&(\mathbf{H}')_{x,y}=\left[\frac{\mathbf{H}+2\pi\mathbf{M}^{NL}}{1-2\pi N\alpha_M}\right]_{x,y}\ .\end{aligned} \tag{32}$$

In the end we come to the following wave equations for electric and magnetic fields:

$$\begin{aligned}
&\nabla\times\nabla\times\mathbf{E}-\mu_z\epsilon k^2\mathbf{E}=4\pi k^2\left[\mu_z\frac{\epsilon+1}{2}\mathbf{P}^{NL}-\mu_z\frac{\epsilon+1}{2}\nabla\cdot\hat{Q}^{NL}+\frac{i}{k}\nabla\times\mathbf{M}^{NL}\right],\\
&(\nabla\times\nabla\times\mathbf{H}-\mu_z\epsilon k^2\mathbf{H})_z=4\pi k^2\left[\epsilon\mathbf{M}^{NL}-\frac{i}{k}\frac{\epsilon+1}{2}\nabla\times\mathbf{P}^{NL}+\frac{i}{k}\frac{\epsilon+1}{2}\nabla\times\nabla\cdot\hat{Q}^{NL}\right]_z,\\
&(\nabla\times\nabla\times\mathbf{E}-\mu\epsilon_z k^2\mathbf{E})_z=4\pi k^2\left[\mu\mathbf{P}^{NL}+\frac{i}{k}\frac{\mu+1}{2}\nabla\times\mathbf{M}^{NL}\right]_z,\\
&\nabla\times\nabla\times\mathbf{H}-\mu\epsilon_z k^2\mathbf{H}=4\pi k^2\left[\epsilon_z\frac{\mu+1}{2}\mathbf{M}^{NL}-\frac{i}{k}\nabla\times\mathbf{P}^{NL}\right].
\end{aligned} \tag{33}$$

The notations are

$$\epsilon\equiv\frac{1+2\pi N\alpha_p+2\pi N\alpha_Q}{1-2\pi N\alpha_p-2\pi N\alpha_Q}\ ,\quad \epsilon_z\equiv 1+4\pi N\alpha_z\ , \tag{34a}$$

$$\mu\equiv\frac{1+2\pi N\alpha_M}{1-2\pi N\alpha_M}\ ,\quad \mu_z\equiv 1+4\pi N(\alpha_M)_z\ . \tag{34b}$$

In the case of $\alpha_Q=0$ we obtain from (34a) the two-dimensional analog of the Lorentz-Lorenz formula

$$\frac{\epsilon-1}{\epsilon+1}=2\pi N\alpha\ . \tag{34c}$$

Thus, like in the three-dimensional case [6], the microscopic parameters $\hat{\alpha},\hat{\alpha}_Q,\hat{\alpha}_M$ reduce to two macroscopic parameters $\hat{\epsilon}$ and $\hat{\mu}$. All quantities that enter into the wave equations for the macroscopic fields are expressible in terms of these two parameters.

In contrast to the three-dimensional case, in planar systems the local field factors f_p and f_Q for electric dipole and electric quadrupole moments are equal: $f_p=f_Q=(\epsilon+1)/2$.

The local field factor f_{2m} for arbitrary m-pole moment in the three-dimensional case is suggested to be

$$f_{2m}=\frac{m(\epsilon+1)+1}{2m+1}\ . \tag{35}$$

From dimensional considerations it may be assumed that for the two-dimensional case this factor takes the form

$$f_{2m}=\frac{m(\epsilon+1)}{2m}=\frac{\epsilon+1}{2}\ . \tag{36}$$

We note that the independence of the factors on multipolarities allows us to significantly simplify the calculation of the medium optical parameters by use of the multipole expansion for an elementary radiator as it will be performed in the following section under investigation of composite media.

The anisotropic, from the three-dimensional viewpoint, character of the problem causes the appearance of the tensors $\hat{\beta}_{M1}$ and $\hat{\eta}_{M2}$, which in the three-dimensional isotropic case equal zero. It is a qualitative distinction. On the other hand, similar to the three-dimensional case, the higher multipole moments are more sensitive to microstructure of the medium. For example, the tensor $\hat{\gamma}$ equals zero both for the quadratic and isotropic lattices, i.e., dipole radiation does not "differentiate" these configurations, whereas the tensor $\hat{\zeta}_1$ turns to zero only for a random arrangement of the cylinders in the xy plane. The calculation of this tensor for a quadratic lattice is presented in Appendix A.

IV. APPLICATION TO COMPOSITE MEDIA AND SELF-ASSEMBLED FILMS

The most evident candidate for the application of the obtained results are quasi-two-dimensional composite media. Take as an example the "mesoscopic" structure when, on the one hand, each column is sufficiently large for forming the macroscopic dielectric permittivity ϵ but, on the other hand, rather small compared with λ and may be considered as an elementary radiator. As justified in the Introduction, if we confine ourselves to considering the dipole radiation only, then the results become applicable to the case of a random distribution of columns in the xy plane. So it is possible to use all the formulas derived for the macroscopic optical parameters of the medium provided that the microscopic polarizability tensor $\hat{\alpha}$ is

$$\alpha_{xx}=\alpha_{yy}=\frac{1}{2}\frac{\epsilon-1}{\epsilon+1}r_0^2\ ,\quad \alpha_{zz}=\frac{\epsilon-1}{4}r_0^2\ . \tag{37}$$

It at once follows from the known solution of the problem about the dielectric cylinder in an external homogeneous field [8]. By use of Eqs. (34) we obtain the following dielectric permittivity tensor ϵ:

$$\begin{aligned}
&\epsilon_{xx}=\epsilon_{yy}=\frac{\epsilon+1+c(\epsilon-1)}{\epsilon+1-c(\epsilon-1)}\ ,\\
&\epsilon_{zz}-1=c(\epsilon-1)\ ,
\end{aligned} \tag{38}$$

where $c=\pi r_0^2N$ is the bulk material concentration.

For a description of the optical properties of the film it is convenient to introduce the optical anisotropy parameter

$$\beta=\frac{\epsilon_{zz}}{\epsilon_{xx}}-1\ . \tag{39}$$

Then from (38) and (39) we obtain

$$\beta=c(1-c)\frac{(\epsilon-1)^2}{\epsilon+1+c(\epsilon-1)}\ . \tag{40}$$

For small concentrations ($c\ll 1$) Eq. (40) coincides with the results of [10], where the influence of the dielectric columns on each other has not been taken into account:

$$\beta=c\frac{(\epsilon-1)^2}{\epsilon+1}\ . \tag{41}$$

For large bulk material concentration $1-c\ll 1$, Eq. (40) takes the form

$$\beta=(1-c)\frac{(\epsilon-1)^2}{2\epsilon} . \quad (42)$$

In both limiting cases the parameter β for homogeneous one-component media naturally turns to zero. The optical anisotropy dependence vs concentration c is shown in Fig. 1, curve I. The maximum value β^{I}_{max} of an anisotropy is

$$\beta^{I}_{max}=(\sqrt{2\epsilon}-\sqrt{\epsilon+1})^2 \quad (43)$$

at concentration

$$c_{max}=\frac{\sqrt{\epsilon+1}}{\epsilon-1}(\sqrt{2\epsilon}-\sqrt{\epsilon+1}) . \quad (44)$$

From (44) it follows that $c_{max}<0.5$ for any magnitude of ϵ. With $\epsilon\to\infty$, c_{max} approaches the upper limit $c_{max}=\sqrt{2}-1\simeq 0.41$. The shift and asymmetry of the $\beta(c)$ dependence are the manifestation of the elongated columnlike morphology of the medium.

Consider such an operation of "phase transformation" when the dielectric regions transform into voids and vice versa. In the general case this means a change of the medium's morphology. But for certain types of inclusion geometry (for example, square rods inserted into a quadratic lattice) the medium's mesostructure does not change. Then the curve $\beta(c)$ will be symmetrical and have a maximum at $c=0.5$.

By use of these results we may calculate the dielectric permittivity tensor of a set of vacuum columns in bulk crystalline silicon. It is the "phase transformed" image of the dielectric columnar mesostructure. Since ϵ is, in fact, the ratio of the dielectric constants, then it is sufficient to substitute $\epsilon\to 1/\epsilon$ and $c\to 1-c$ in Eq. (38). Then instead of Eq. (38) we get

$$\epsilon_{xx}=\epsilon_{yy}=\epsilon\frac{2+c(\epsilon-1)}{2\epsilon-c(\epsilon-1)} , \quad \epsilon_{zz}-1=c(\epsilon-1) , \quad (45)$$

where c is the concentration of crystalline silicon.

Curve II in Fig. 1 shows the $\beta(c)$ dependence for vacuum columns in the bulk dielectric structures. This dependence strongly differs from that given by curve I. The maximum value of the anisotropy β^{II}_{max} equals

$$\beta^{II}_{max}=\frac{1}{\epsilon}\left[\frac{\sqrt{\epsilon+1}+\sqrt{2\epsilon}}{\sqrt{\epsilon+1}+\sqrt{2}}\right]^2\beta^{I}_{max} \quad (46)$$

and for large $\epsilon\gg 1$ this value is about ϵ times less than that for the silicon column-type geometry:

$$1-c_{max}=\frac{\sqrt{\epsilon+1}}{\epsilon-1}(\sqrt{\epsilon+1}-\sqrt{2}) \quad (47)$$

This example with dielectric and vacuum columns vividly demonstrates that the macroscopic optical properties of the composite system may dramatically depend not only on the concentration of the inclusions but *on their form as well.*

Nonlinear susceptibility presents an even more striking example. Since the field acting on the cylinder multiplies the macroscopic field by a factor of $(\epsilon_{xx}+1)/2$, then for the third-order susceptibility, with allowance for Eq. (36), we have a factor of $[(\epsilon_{xx}+1)/2]^4$. For the mesostructure of the silicon cylinders the field inside each cylinder is less than the acting field by a factor of $(\epsilon+1)/2$ [8], whereas for the mesostructure of the vacuum cylinders this factor is of the order of unity. As a result, for the silicon and vacuum columnar mesostructures the nonlinear susceptibilities would differ by two orders of magnitude.

We would like to emphasize once more that the consideration of columnlike porous media was carried out under the assumption of purely dipole fields from the columns. This holds true either for arbitrary relative positions of the columns when the intercolumn distances are large in comparison with their diameters or for arbitrary concentrations under a random disposition of the columns. The case of the ordered locations *and* the very high concentrations demands that the further multipole moments of the radiator's field be taken into account. This problem will be considered elsewhere.

With regard to the optical properties of LB films [1,11,12] and organic films in general [13], this is motivated by the high nonlinearity of the organic molecules which compose the films. Equations (22)–(26) may be applied immediately for calculation of these macroscopic optical properties, i.e., linear and nonlinear susceptibilities, if the polarizabilities of the molecules are known. Previously, calculations of these nonlinear susceptibilities were performed with a customary three-dimensional local field factor $(\epsilon+2)/3$ [14]. But for LB and self-assembled films constructed from elongated molecular chains it is much more reasonable to use the 2D factor $(\epsilon+1)/2$. Even for moderate values of $\epsilon\sim 4$ the difference between these factors is about 15–25 %. Since the second- and third-order susceptibilities $\chi^{(2)}$ and $\chi^{(3)}$ are proportional to the third and fourth powers of the local field factor, then the distinction between the calculations with two- or three-dimensional local-field factors may reach 100% or more. The account of the regular properties of 2D structures [see Eqs. (24)–(26)] gives a still more vivid distinction from the usual isotropic 3D approach. Sufficiently exact measurements of the polarizabilities and susceptibilities will allow us, with the help of Eqs. (10), (11), and (23)–(26), to obtain information about molecular spatial configurations in the film. Valid information about local-

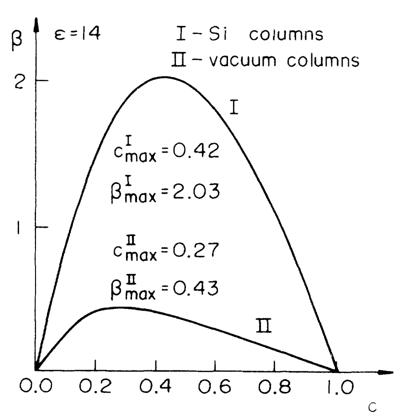

FIG. 1. Anisotropy parameter β as a function of silicon concentration c. I, silicon columns in the vacuum; II, vacuum columns in bulk silicon.

field factors may be essential for optimization of the film's parameters [15].

V. SUMMARY

The general idea of the substitution of variables in the MIEs happens to be effective for the analysis of quasi-two-dimensional optical media. Just as in the three-dimensional case the optical parameters of the medium contain information about the geometry of the mesostructure with the characteristic sizes much less than the wavelength. Moreover, the finer peculiarities of such a mesostructure are described by the multipole moments of higher order. For example, the tensor ζ_1 of the "quadrupolar origin" equals zero for the chaotic medium in the xy plane and differs from zero for the quadratic lattice whereas the "dipole tensor" $\hat{\gamma}\equiv 0$ in both cases. The second example presents a quasi-two-dimensional medium with random isotropic arrangement of the radiators (cylinders) in the xy plane: in such a system the tensors of the magneto-dipole origin $\hat{\eta}_M$ and $\hat{\eta}_{M2}$ differ from zero whereas for really isotropic random three-dimensional media these tensors are exactly equal to zero.

After the substitution of the three-dimensional Green's function by the two-dimensional one and the replacement of the surface integration by integration along the boundary lines, an extinction theorem for the two-dimensional medium coincides with its three-dimensional analog.

The concept of the elementary cylinder radiator (mesoscopic atom) converts the MIE into quite an effective tool for the analysis of optical properties of composite media, while for the case of the random (three- or two-dimensional) distribution of inclusions the results obtained through the dipole approximation may be directly applied to composite media. In particular, it may be argued that the three-dimensional Maxwell-Garnett formula

$$\epsilon-1=\frac{4\pi N\alpha}{1-\frac{4\pi}{3}N\alpha} \tag{48}$$

and the formula for the mesoscopic polarizability α

$$\alpha=\frac{\epsilon-1}{\epsilon+2}r_0^3\ ,\quad \text{i.e.,}\quad \epsilon=\frac{\epsilon+2+2c(\epsilon-1)}{\epsilon+2-c(\epsilon-1)}\ , \tag{49}$$

obtained on its basis for the spherical inclusions, hold true for arbitrary concentrations c of the inclusions. Under the regular arrangement of the inclusions and high concentrations we must keep the multipolar terms.

As well as in the three-dimensional case [6] calculations in Appendixes A and B and all the rest of our calculations are made with an accuracy up to the parameter ka inclusive. Therefore, all these results hold true with the same accuracy not only in the limit $kb\rightarrow 0$ but also in the first-order approximation. Consideration of the retention of higher-order terms [in this case the terms $\sim(kb)^2\ln(kb)$] will be given elsewhere. The technique developed, in our opinion, is the most useful for the analysis of composite media with inclusion sizes and interinclusion distances not too small compared to the wavelength, just as in the case of the workable two-dimensional PBG structures.

After our work was submitted for publication we became aware of the recently published paper by Robertson *et al.* [16] about the measurement of the dielectric constants for different orthogonal polarizations in two-dimensional order dielectric arrays. In the limiting case of a long wavelength they obtained good agreement with the phenomenological Weiner theory for an E field parallel to the rods, but for the perpendicular E field there is strong disagreement, by a factor of about 2, for the refractive index $(n_\perp-1)$ (experimentally equal to 0.11 and 0.09 for quadratic and triangular lattices, respectively) in comparison with the prediction of the phenomenological theory of 0.06 and 0.05, respectively [Eq. (6) of Ref. [16]]. Substitution of their experimental concentrations c for both types of lattices and the dielectric constant ϵ of the material ($c=0.125$, 0.11, and $\epsilon=8.6$) into our Eq. (38) immediately gives for the lattices' refractive index $(n_\perp-1)$ the values 0.11 and 0.091, respectively, in splendid agreement with the cited experiment. This correspondence is even better than one would expect because our theory takes into account only the first (dipole) term of the multipole expansion of the elementary radiator's field. However, one could object that the field of a dielectric cylinder has pure dipole form only in a uniform external field. Such a situation exists within the random disposition of radiators and therefore each cylinder behaves as an ideal two-dimensional dipole for arbitrary concentrations of the inclusions. It seems that symmetry of the triangular and quadratic lattice is sufficient to ensure the quasiuniformity of the field in the vicinity of lattice points.

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APPENDIX A: THE CALCULATION OF TENSORS $\hat{\gamma}$ AND $\hat{\zeta}$ FOR THE REGULAR SQUARE LATTICE AND CHAOTIC MEDIA

From a requirement of the tensor equality covariance for the central symmetrical media we get the condition

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

For the calculation of the tensors $\hat{\gamma}$, $\hat{\zeta}_1$, and $\hat{\zeta}_M$ the general idea based on the fact that for the medium with two orthogonal axes of symmetry (rectangular lattice) all the terms with odd powers of the Cartesian components $\mathbf{n}_{lj}$ turn to zero,

$$(\hat{\gamma})_{st}=2b^2\sum_j^{\sigma}\frac{2(\mathbf{n}_{lj})_s^2+\delta_{sz}-1}{R_{lj}^2}\delta_{st}\ , \tag{A2}$$

$$(\hat{\zeta}_1)_{stpq}=8b^2\sum_j^{\sigma}[(1-\delta_{sz})\delta_{sq}\delta_{tp}(\mathbf{n}_{lj})_t^2 -2(2\delta_{sq}\delta_{tp}+\delta_{st}\delta_{pq})(1-\tfrac{2}{3}\delta_{sp}) \times(\mathbf{n}_{lj})_s^2(\mathbf{n}_{lj})_p^2]\frac{1}{R_{lj}^2}\ , \tag{A3}$$

and

$$(\hat{\zeta}_M)_{stp}=2b^2\sum_j^{\sigma}\frac{2(\mathbf{n}_{lj})_t^2+\delta_{tz}}{R_{lj}^2}\epsilon_{stp}\ . \tag{A4}$$

For the square lattice $\hat{\gamma}\equiv 0$, $\hat{\zeta}_M\sim\hat{\epsilon}$ and the convolution $\hat{\zeta}_M$ with the symmetric tensor $\hat{Q}$ equals zero. Therefore, we may put into (10) the tensor $\hat{\zeta}_M=0$ and write the expression for $\hat{\zeta}_1$ in the following form [6]:

$$(\hat{\zeta}_1)_{ssss}|_{s\neq z}=-(\hat{\zeta}_1)_{stts}|_{s\neq t,s\neq z} =4b^2\sum\frac{3-8(n_{lj})_s^4}{R_{lj}^2}=9.04\ , \tag{A5a}$$

$$(\hat{\zeta}_1)_{ztpq}=0 \tag{A5b}$$

whereas the rest of the components of the tensor $\hat{\zeta}_1$ equal zero.

Finally, for a chaotic distribution of the radiators, taking advantage of the averaging over an ensemble of the random spatial configurations, summation may be changed for integration. All zeroth tensor components of the square lattice will equal zero after the averaging. But, besides, due to the angular dependence $(\mathbf{n}_{je})_s$, the right-hand side of Eq. (A5a) after angular integration turns to zero. As a result one obtains Eq. (12).

APPENDIX B: THE FACTORING OF THE OPERATOR ∇×∇× OUTSIDE THE INTEGRAL SIGN

We start from an equality

$$\frac{\partial}{\partial x_i}\int_\sigma^\Sigma F d^2\mathbf{r}'=\int_\sigma^\Sigma\frac{\partial F}{\partial x_i}d^2\mathbf{r}'+\int_\sigma F(\mathbf{n})_i d\mathbf{r}'\ , \tag{B1}$$

where $F=F(\mathbf{r},\mathbf{r}')$ is an arbitrary function and $\mathbf{n}$ is the unit vector normal to the boundary σ. Set $F=i\pi H_0^{(1)}(kR)f(\mathbf{r}')$ and, taking into account the small size of a, expand $f(\mathbf{r}')$ in a power series in the vicinity of the point $\mathbf{r}$:

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

Calculating directly integrals over σ in the right-hand side of the equality (B1) we obtain

$$\int_\sigma fGn_i dr'=i\pi aH_0^{(1)}(ka)f\int n_i d\Omega-i\pi a^2H_0^{(1)}(ka)\frac{\partial f}{\partial x_j}\int n_i n_j d\Omega+\tfrac{1}{2}i\pi a^3H_0^{(1)}(ka)\frac{\partial^2 f}{\partial x_j\partial x_k}\int n_i n_j n_k d\Omega +O\left[\frac{\partial^4 f}{\partial x^4}a^4\ln(ka)\right]=-i\pi^2a^2H_0^{(1)}\frac{\partial f}{\partial x_i}+O\left[\frac{\partial^4 f}{\partial x^4}a^4\ln(ka)\right]=O+O[a^2\ln(ka)]\ , \tag{B3a}$$

$$\int_\sigma f\frac{\partial G}{\partial x_j}n_i dr'=-2\pi f\delta_{ij}(1-\delta_{iz})+O(a^2\ln a)\ , \tag{B3b}$$

$$\int_\sigma f\frac{\partial^2 G}{\partial x_j\partial x_k}n_i dr'=\pi\left[\delta_{jk}\frac{\partial f}{\partial x_i}-\delta_{ij}\frac{\partial f}{\partial x_k}-\delta_{ik}\frac{\partial f}{\partial x_j}\right](1-\delta_{iz})(1-\delta_{jz})+O[a^2\ln(ka)]\ , \tag{B3c}$$

$$\int_\sigma f\frac{\partial^3 G}{\partial x_j\partial x_k\partial x_e}n_i dr'=\pi\left\{\frac{1}{2}\left[k^2f+\frac{1}{3}\frac{\partial^2 f}{\partial x_m^2}\right](\delta_{ij}\delta_{ke}+\delta_{ik}\delta_{je}+\delta_{ie}\delta_{jk})(1-\delta_{iz})(1-\delta_{jz})(1-\delta_{kz}) +\frac{1}{3}\left[\frac{\partial^2 f}{\partial x_i\partial x_j}\delta_{ke}+\frac{\partial^2 f}{\partial x_i\partial x_k}\delta_{jl}+\frac{\partial^2 f}{\partial x_i\partial x_e}\delta_{jk}\right](1-\delta_{jz})(1-\delta_{kz}) -\frac{2}{3}\left[\frac{\partial^2 f}{\partial x_k\partial x_e}\delta_{ij}+\frac{\partial^2 f}{\partial x_j\partial x_e}\delta_{ik}+\frac{\partial^2 f}{\partial x_j\partial x_k}\delta_{ie}\right](1-\delta_{iz})\right\}\ . \tag{B3d}$$

Formulas (B3) are obtained with the accuracy up to the first-order terms in ka inclusive. By use of Eqs. (B1) and (B3) we get the equalities with the same accuracy up to the terms ka inclusive:

$$\nabla\times\int_\sigma^\Sigma \mathbf{f}G\,d^2\mathbf{r}'=\int_\sigma^\Sigma\nabla\times\mathbf{f}G\,d^2\mathbf{r}'\ , \tag{B4a}$$

$$\left[\nabla\times\int_\sigma^\Sigma\nabla\times\mathbf{f}G\,d^2\mathbf{r}'\right]_i =\left[\int_\sigma^\Sigma\nabla\times\nabla\times\mathbf{f}G\,d^2\mathbf{r}'\right]_i+2\pi(1+\delta_{iz})(\mathbf{f})_i\ , \tag{B4b}$$

$$\left[\nabla\times\int_\sigma^\Sigma\nabla\times\nabla\times\mathbf{f}G\,d^2\mathbf{r}'\right]_i =\left[\int_\sigma^\Sigma\nabla\times\nabla\times\nabla\times\mathbf{f}G\,d^2\mathbf{r}'\right]_i+2\pi\delta_{iz}(\nabla\times\hat{f})_z\ , \tag{B4c}$$

$$\left[\nabla\times\int_\sigma^\Sigma\nabla\cdot\hat{f}G\,d^2\mathbf{r}'\right]_i =\left[\int_\sigma^\Sigma\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}'\right]_i-2\pi(1-\delta_{jz})\epsilon_{ijk}(\hat{f})_{jk}\ , \tag{B4d}$$

$$\left[\nabla\times\int_\sigma^\Sigma\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}'\right]_i = \left[\int_\sigma^\Sigma\nabla\times\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}'\right]_i + 2\pi(\nabla\cdot\hat{f})_i + \pi(1-\delta_{iz})[\nabla\cdot(\hat{f}^*-\hat{f})-\nabla\,\mathrm{Tr}\hat{f}]_i\ , \quad \text{(B4e)}$$

$$\left[\nabla\times\int_\sigma^\Sigma\nabla\times\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}'\right]_i = \left[\int_\sigma^\Sigma\nabla\times\nabla\times\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}'\right]_i + 2\pi\left[\epsilon_{ijk}(\delta_{jz}-1)k^2f_{jk} + \delta_{iz}\left[\nabla\times\nabla\cdot\frac{\hat{f}+\hat{f}^*}{2}\right]_i\right]\ . \quad \text{(B4f)}$$

Repeatedly using Eqs. (B4) we obtain the desired relationships:

$$\nabla\times\nabla\times\int_\sigma^\Sigma\mathbf{f}G\,d^2\mathbf{r}' = \int_\sigma^\Sigma\nabla\times\nabla\times\mathbf{f}G\,d^2\mathbf{r}' + \hat{\Psi}\cdot\mathbf{f}\ ,$$
$$(\hat{\Psi})_{st} = 2\pi\delta_{st}(1+\delta_{sz})\ ; \quad \text{(B5a)}$$

$$\nabla\times\nabla\times\int_\sigma^\Sigma\nabla\times\mathbf{f}G\,d^2\mathbf{r}' = \int_\sigma^\Sigma\nabla\times\nabla\times\nabla\times\mathbf{f}G\,d^2\mathbf{r}' + \hat{\Psi}_{11}:(\nabla\mathbf{f})\ , \quad \text{(B5b)}$$
$$(\hat{\Psi}_{11})_{stk} = 4\pi\epsilon_{stk}\ ;$$

$$\nabla\times\nabla\times\int_\sigma^\Sigma\nabla\cdot\hat{f}G\,d^2\mathbf{r}' = \int_\sigma^\Sigma\nabla\times\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}' + \hat{\Theta}_{11}\vdots(\nabla\hat{f})\ ,$$
$$(\hat{\Theta}_{11})_{stkp} = 4\pi\delta_{sp}\delta_{tk} + \pi(\delta_{sz}-1)(\delta_{sp}\delta_{tk}+\delta_{sk}\delta_{tp}+\delta_{st}\delta_{kp})\ ; \quad \text{(B5c)}$$

$$\nabla\times\nabla\times\int_\sigma^\Sigma\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}' = \int_\sigma^\Sigma\nabla\times\nabla\times\nabla\times\nabla\cdot\hat{f}G\,d^2\mathbf{r}' + k^2\hat{\Theta}_2:\hat{f} + \hat{\Theta}_{22}\vdots(\nabla\nabla\hat{f})\ ,$$
$$(\hat{\Theta}_{22})_{stkpq} = 2\pi\epsilon_{stq}\delta_{kp} + 2\pi\epsilon_{stp}\delta_{kq}\delta_{sz}\ ,$$
$$(\hat{\Theta}_2)_{stk} = 2\pi\epsilon_{stk}(\delta_{tz}-1)\ . \quad \text{(B5d)}$$

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