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RELATION BETWEEN THE NONLINEAR DIELECTRIC CONSTANT AND THE GREEN'S FUNCTIONS FOR ELECTROMAGNETIC RADIATION

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A formula is established expressing the nonlinear dielectric permittivity tensor $\beta_{ijk}(\mathbf{k}, \mathbf{k}'; \omega, \omega')$, which describes effects of second order in the electromagnetic field, in terms of the triple-time retarded Green's functions for electromagnetic radiation in matter. Derivation of the formula is based on application of the method of external currents and perturbation theory for the density matrix of the system.

1. As is well known,[1,2] in linear electrodynamics there is a definite relation between the Green's functions for electromagnetic radiation in a medium and its dielectric constant. In the present article we consider the case of an anisotropic medium whose interaction with electromagnetic radiation is weakly nonlinear[3] and derive formulas expressing the nonlinear terms of the dielectric permittivity in terms of retarded Green's functions for the radiation in the medium. In this connection we confine our attention to nonlinear effects of second order in the field. The medium is assumed to be homogeneous and nonmagnetic. The Gaussian system of units is used, in which we set $\hbar = c = 1$.

2. As follows from the principle of causality, a nonlinear local relation between the displacement **D** and the intensity **E** of the electromagnetic field in a medium may be represented in the form

$$D_i(\mathbf{r}, t) = E_i(\mathbf{r}, t) + \int_0^\infty f_{ij}(\tau) E_j(\mathbf{r}, t-\tau)\, d\tau + \int_0^\infty\int_0^\infty f_{ijk}(\tau_1, \tau_2) E_j(\mathbf{r}, t-\tau_1) E_k(\mathbf{r}, t-\tau_2)\, d\tau_1\, d\tau_2 + \ldots,$$

or for the Fourier component with respect to the time

$$D_i(\mathbf{r}, \omega) = E_i(\mathbf{r}, \omega) + \beta_{ij}(\omega) E_j(\mathbf{r}, \omega) + \int_{-\infty}^{\infty} \beta_{ijk}(\omega_1, \omega-\omega_1) E_j(\mathbf{r}, \omega_1) E_k(\mathbf{r}, \omega-\omega_1)\, d\omega_1 + \ldots, \tag{1}$$

where

$$\beta_{ij}(\omega) = \int_0^\infty f_{ij}(\tau) e^{i\omega\tau}\, d\tau,$$

$$\beta_{ijk}(\omega_1, \omega_2) = \int_0^\infty\int_0^\infty \exp\{i(\omega_1\tau_1 + \omega_2\tau_2)\} f_{ijk}(\tau_1, \tau_2)\, d\tau_1\, d\tau_2, \ldots \tag{2}$$

In what follows, we assume that the terms in (1) which are nonlinear in the field are small.

We shall use the method of given external currents $\mathbf{j}^{\text{ext}}(\mathbf{r}, \omega)$.[1] Eliminating the magnetic field from the system of macroscopic Maxwell's equations, we obtain

$$\operatorname{rot}\operatorname{rot}\mathbf{E}(\mathbf{r}, \omega) - \omega^2\mathbf{D}(\mathbf{r}, \omega) = 4\pi i\omega\mathbf{j}^{\text{ext}}(\mathbf{r}, \omega). \tag{3*}$$

In what follows, we omit superscript "ext" on **j**.

In accordance with (1), we seek the solution of Eq. (3) in the form of an expansion

$$\mathbf{E}(\mathbf{r}, \omega) = \mathbf{E}^{(1)}(\mathbf{r}, \omega) + \mathbf{E}^{(2)}(\mathbf{r}, \omega) + \ldots$$

By the method of iterations we obtain

$$[(\operatorname{rot}\operatorname{rot})_{ij} - \omega^2\varepsilon_{ij}(\omega)] E_j^{(1)}(\mathbf{r}, \omega) = 4\pi i\omega j_i(\mathbf{r}, \omega), \tag{4}$$

$$[(\operatorname{rot}\operatorname{rot})_{ij} - \omega^2\varepsilon_{ij}(\omega)] E_j^{(2)}(\mathbf{r}, \omega) = \omega^2 \int d\omega_1\, \beta_{ijk}(\omega_1, \omega-\omega_1) E_j^{(1)}(\mathbf{r}, \omega_1) E_k^{(1)}(\mathbf{r}, \omega-\omega_1), \ldots; \tag{5}$$

where

$$\varepsilon_{ij}(\omega) = \delta_{ij} + \beta_{ij}(\omega), \quad \operatorname{rot}\operatorname{rot}_{ij} E_j \equiv (\operatorname{rot}\operatorname{rot}\mathbf{E})_i.$$

One can write down the solution by introducing the Green's function of Eq. (4), $G_{ik}(\mathbf{r}, \mathbf{r}_1, \omega)$,[1] which satisfies the following equation:

$$[(\operatorname{rot}\operatorname{rot})_{ij} - \omega^2\varepsilon_{ij}] G_{jl}(\mathbf{r}, \mathbf{r}_1; \omega) = \delta_{il}\delta(\mathbf{r}-\mathbf{r}_1). \tag{6}$$

With the aid of this function we have

$$E_i^{(1)}(\mathbf{r}, \omega) = 4\pi i\omega \int d^3r_1 G_{il}(\mathbf{r}, \mathbf{r}_1; \omega) j_l(r_1, \omega), \tag{7}$$

$$E_i^{(2)}(\mathbf{r}, \omega) = \omega^2 \int d^3r_1\, G_{ij}(\mathbf{r}, \mathbf{r}_1; \omega) \int_{-\infty}^{\infty} d\omega_1\, \beta_{jkl}(\omega_1, \omega-\omega_1) \times E_k^{(1)}(\mathbf{r}_1, \omega_1) E_l^{(1)}(\mathbf{r}_1, \omega-\omega_1), \ldots \tag{8}$$

*rot ≡ curl.

Then changing to a Fourier representation with respect to the spatial coordinates, taking into consideration that in a homogeneous medium G_{ij} only depends on the difference $\mathbf{r} - \mathbf{r}_1$, and also substituting (7) into (8), we obtain

$$E_i^{(1)}(\mathbf{k}, \omega) = 4\pi i\omega(2\pi)^3 G_{il}(\mathbf{k}, \omega) j_l(\mathbf{k}, \omega), \quad (9)$$

$$E_i^{(2)}(\mathbf{k}, \omega) = (4\pi i\omega)^2 (2\pi)^9 \int_{-\infty}^{\infty} d\omega_1\, \omega_1(\omega - \omega_1)\beta_{jkl}(\omega_1, \omega - \omega_1) \times \int d^3k_1\, G_{ij}(\mathbf{k}, \omega) G_{ks}(\mathbf{k}_1, \omega_1) \times G_{lp}(\mathbf{k}-\mathbf{k}_1, \omega-\omega_1) j_s(\mathbf{k}_1, \omega_1) j_p(\mathbf{k}-\mathbf{k}_1, \omega-\omega_1), \ldots \quad (10)$$

3. On the other hand, we can express the macroscopic fields $\mathbf{E}^{(1)}, \mathbf{E}^{(2)}, \ldots$ in terms of microscopic parameters with the aid of statistical averaging. In the presence of external currents, the Hamiltonian H_0 of the system acquires the additional term

$$\hat{V}(t) = -\int d^3r \hat{A}_\nu(\mathbf{r}) j_\nu(\mathbf{r}, t).$$

Here

$$\hat{A}_\nu(\mathbf{r}) = (\hat{\mathbf{A}}(\mathbf{r}), i\hat{\varphi}(\mathbf{r}))$$

is the Schrödinger operator of the 4-vector potentials,

$$j_\nu(\mathbf{r}, t) = (\mathbf{j}(\mathbf{r}, t), i\rho(\mathbf{r}, t))$$

is the 4-vector of the external currents. Here and in what follows, Greek indices label the components of 4-tensors, but Latin indices pertain to their spatial part; everywhere, summation over repeated indices is to be understood.

One can find the macroscopic average of the 4-vector potential from the formula

$$A_\nu(\mathbf{r}, t) = \mathrm{Sp}\,(\hat{\rho}'(t)\, \hat{A}_\nu(\mathbf{r}, t)),$$

where ρ' is the density matrix, and also it is convenient to use the interaction representation in which

$$\hat{A}_\nu(\mathbf{r}, t) = e^{iH_0t} \hat{A}_\nu(\mathbf{r}) e^{-iH_0t},$$

and ρ' satisfies the equation

$$\frac{\partial \hat{\rho}'(t)}{\partial t} = -i\,[\hat{U}(t), \hat{\rho}'(t)], \qquad \hat{U} = e^{iH_0t}\hat{V}e^{-iH_0t}.$$

The solution of this equation, obtained by the method of successive approximations, has the form [4]

$$\hat{\rho}' = \hat{\rho}^{(0)} + \hat{\rho}^{(1)} + \hat{\rho}^{(2)} + \ldots,$$

where

$$\hat{\rho}^{(n)} = (-i)^n \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \ldots \times \int_{-\infty}^{t_{n-1}} dt_n\, [\hat{U}(t_1)[\ldots[\hat{U}(t_n)\hat{\rho}]\ldots];$$

here $\hat{\rho} = \hat{\rho}(0)$ is the density matrix of the system, unperturbed by an external current. Correspondingly

$$A_\nu = A_\nu^{(0)} + A_\nu^{(1)} + A_\nu^{(2)} + \ldots, \qquad A_\nu^{(n)} = \mathrm{Sp}\,(\rho'^{(n)}\hat{A}_\nu).$$

In particular,

$$A_\nu^{(1)}(\mathbf{r}, t) = -\int_{-\infty}^{\infty} dt_1 \int d^3r_1\, \mathscr{D}_{\nu\mu}^R(\mathbf{r}, t; \mathbf{r}_1, t_1) j_\mu(\mathbf{r}_1, t_1), \quad (11)$$

where $\mathscr{D}_{\nu\mu}^R(\mathbf{r}, t; \mathbf{r}_1, t_1)$ is the usual double-time retarded Green's function for the electromagnetic field.[1] Then it is not difficult to verify that

$$A_\nu^{(2)}(\mathbf{r}, t) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \times \int\int d^3r_1 d^3r_2\, \mathscr{D}_{\nu\mu\tau}^R(\mathbf{r}, t; \mathbf{r}_1, t_1; \mathbf{r}_2, t_2) j_\mu(\mathbf{r}_1, t_1) j_\tau(\mathbf{r}_2, t_2), \quad (12)$$

where the triple-time retarded Green's function $\mathscr{D}_{\nu\mu\tau}^R$ is determined by the following expression:

$$\mathscr{D}_{\nu\mu\tau}^R(\mathbf{r}, t; \mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = -\theta(t - t_1)\theta(t_1 - t_2) \times \mathrm{Sp}\,\{\hat{\rho}[[\hat{A}_\nu(\mathbf{r}, t), \hat{A}_\mu(\mathbf{r}_1, t_1)]\hat{A}_\tau(\mathbf{r}_2, t_2)]\} \quad (13)$$

($\theta(t) = 1$ for $t > 0$ and $\theta(t) = 0$ for $t < 0$).

The function $\mathscr{D}_{\nu\mu\tau}^R$ given by expression (13) only depends on the difference of time ($t - t_1$, $t - t_2$) and, in a homogeneous medium—on the difference of the spatial arguments. Taking this fact into consideration, changing to the Fourier representation in (11) and (12)

$$\mathscr{D}_{\nu\mu}^R(\mathbf{r}, t) = \int d^3k \int d\omega\, \mathscr{D}_{\nu\mu}^R(\mathbf{k}, \omega) \exp\{i(\mathbf{kr} - \omega t)\}$$

and other equations, we obtain

$$A_\nu^{(1)}(\mathbf{k}, \omega) = -(2\pi)^4 \mathscr{D}_{\nu\mu}^R(\mathbf{k}, \omega) j_\mu(\mathbf{k}, \omega), \quad (14)$$

$$A_\nu^{(2)}(\mathbf{k}, \omega) = (2\pi)^8 \int d\omega' \int d^3k'\, \mathscr{D}_{\nu\mu\tau}^R(\mathbf{k}', \mathbf{k} - \mathbf{k}'; \omega', \omega - \omega') \times j_\mu(\mathbf{k}'\omega') j_\tau(\mathbf{k} - \mathbf{k}'; \omega - \omega'). \quad (15)$$

Now let us express the field E_j in terms of the retarded Green's functions $\mathscr{D}^R$ by using the fact that

$$E_j^{(n)}(\mathbf{r}, t) = -\frac{\partial}{\partial t} A_j^{(n)}(\mathbf{r}, t) - \nabla_j \varphi^{(n)}(\mathbf{r}, t)$$

or

$$E_j^{(n)}(\mathbf{k}, \omega) = i\omega A_j^{(n)}(\mathbf{k}, \omega) - k_j A_0^{(n)}(\mathbf{k}, \omega).$$

In particular,

$$E_j^{(1)}(\mathbf{k}, \omega) = -(2\pi)^4 \{[i\omega \mathscr{D}_{jl}^R(\mathbf{k}, \omega) - k_j \mathscr{D}_{0l}^R(\mathbf{k}, \omega)] j_l(\mathbf{k}, \omega) + [i\omega \mathscr{D}_{j0}^R(\mathbf{k}, \omega) - k_j \mathscr{D}_{00}^R(\mathbf{k}, \omega)] j_0(\mathbf{k}, \omega)\}. \quad (16)$$

Considering the equation of continuity, $\mathrm{div}\, \mathbf{j} + (\partial\rho/\partial t) = 0$ or $i(\mathbf{k}\cdot\mathbf{j}) - \omega j_0 = 0$, we write Eq. (16) in the form

$$E_j^{(1)}(\mathbf{k}, \omega) = 4\pi i\omega(2\pi)^3 \mathscr{D}_{jl}^E(\mathbf{k}, \omega) j_l(\mathbf{k}, \omega); \quad (17)$$

$$\mathscr{D}_{jl}^E(\mathbf{k},\omega) = -S_{jl}(\mathbf{k},\omega) - \frac{ik_l}{\omega} S_{j0}(\mathbf{k},\omega),$$

$$S_{jl} = \frac{1}{2}\mathscr{D}_{jl}^R + \frac{ik_j}{\omega}\mathscr{D}_{0l}^R, \quad S_{j0} = \frac{1}{2}\mathscr{D}_{j0}^R + \frac{ik_j}{\omega}\mathscr{D}_{00}^R. \tag{18}$$

Similarly we can verify that

$$E_j^{(2)}(\mathbf{k},\omega) = (4\pi i\omega)^2(2\pi)^9 \int d\omega' \times \int d^3k' \, \mathscr{D}^E_{jmn}(\mathbf{k}', \mathbf{k}-\mathbf{k}'; \omega', \omega-\omega') \times j_m(\mathbf{k}',\omega') j_n(\mathbf{k}-\mathbf{k}', \omega-\omega'); \tag{19}$$

$$\mathscr{D}^E_{jmn} = \frac{1}{4\pi i\omega(2\pi)^2}\left[S_{jmn} + \frac{ik_m'}{\omega'} S_{j0n} + \frac{i(k_n - k_n')}{\omega-\omega'} S_{jm0} - \frac{k_m'(k_n-k_n')}{\omega'(\omega-\omega')} S_{j00}\right],$$

$$S_{jmn} = \frac{1}{2}\mathscr{D}^R_{jmn} + \frac{ik_j}{2\omega}\mathscr{D}^R_{0mn}, \quad S_{j0n} = \frac{1}{2}\mathscr{D}^R_{j0n} + \frac{ik_j}{2\omega}\mathscr{D}_{00n} \tag{20}$$

and similarly for S_{jm0} and S_{j00}. All functions $\mathscr{D}$ and S in Eq. (20) have $(\mathbf{k}', \mathbf{k} - \mathbf{k}'; \omega', \omega - \omega')$ as their arguments.

Comparison of (17) and (19) with (9) and (10) enables us, in virtue of the arbitrariness of the external currents, to identify the functions $\mathscr{D}^E_{ij}$ and G_{ij}, and also to establish the fact that

$$\mathscr{D}^E_{jmn}(\mathbf{k}', \mathbf{k}-\mathbf{k}'; \omega', \omega-\omega') = \omega'(\omega-\omega')\beta_{kls}(\omega', \omega-\omega') \times \mathscr{D}_{jk}^E(\mathbf{k},\omega)\mathscr{D}_{lm}^E(\mathbf{k}',\omega')\mathscr{D}_{sn}^E(\mathbf{k}-\mathbf{k}', \omega-\omega'). \tag{21}$$

As follows from (6), the functions $\mathscr{D}^E_{jk}$ satisfy the equation

$$B_{ij}(\mathbf{k},\omega)\mathscr{D}_{jl}^E(\mathbf{k},\omega) = \delta_{il}, \quad B_{ij} = k^2\delta_{ij} - k_ik_j - \omega^2\varepsilon_{ij}(\omega). \tag{22}$$

As is evident from (22), the tensor B_{ij} is symmetric; therefore the tensor $\mathscr{D}^E_{ij}$ also is symmetric.

Multiplying both sides of (21) by

$$B_{ij}(\mathbf{k},\omega)B_{pm}(\mathbf{k}',\omega')B_{rn}(\mathbf{k}-\mathbf{k}', \omega-\omega')$$

and summing over j, m, and n, it is not difficult to obtain the desired expression for β_{ijk}:

$$\beta_{ijk}(\omega,\omega') = (\omega\omega')^{-1}B_{il}(\mathbf{k}+\mathbf{k}', \omega+\omega') \times B_{jm}(\mathbf{k},\omega)B_{kn}(\mathbf{k}',\omega')\,\mathscr{D}^E_{lmn}(\mathbf{k},\mathbf{k}';\omega,\omega'). \tag{23}$$

Formula (23) is valid for arbitrary $\mathbf{k}$ and $\mathbf{k}'$. In particular, one can set $\mathbf{k} = \mathbf{k}' = 0$; then

$$\beta_{ijk}(\omega,\omega') = \frac{i(\omega+\omega')\omega\omega'}{4(2\pi)^3}\varepsilon_{il}(\omega+\omega') \times \varepsilon_{jm}(\omega)\varepsilon_{kn}(\omega')\mathscr{D}^R_{lmn}(\omega,\omega'). \tag{24}$$

For the sake of simplicity, we did not consider spatial dispersion [5] of the dielectric constant; however, generalization of the theory to this case is not difficult. Being given from the very beginning a nonlocal relation between **D** and **E**, it is necessary to also change in (1) to a Fourier representation with respect to the spatial coordinates, having written the nonlinear term in the form

$$\int \beta_{ijk}(\mathbf{k}', \mathbf{k}-\mathbf{k}'; \omega', \omega-\omega')E_j(\mathbf{k}',\omega') \times E_k(\mathbf{k}-\mathbf{k}'; \omega-\omega')\,d^3k'd\omega'.$$

It is not difficult to verify that the final formula (23) remains in force if $\beta_{ijk}(\mathbf{k}, \mathbf{k}'; \omega, \omega')$ is substituted on the left.

Thus, the nonlinear dielectric tensor is expressed in terms of the linear approximation for the dielectric constants and the Fourier components of the triple-time retarded Green's functions for electromagnetic radiation in a medium. This enables one to apply the methods of quantum field theory in order to study the propagation of electromagnetic waves in media possessing nonlinear properties. Similar methods may also be applied to the higher-order approximations.

For various special choices of potential gauge, the general formula (20) determining D^E_{ijk} may be simplified. Thus, in the case of the gauge $\varphi = 0$, only the spatial components of $D^R_{\alpha\gamma\beta}$ and of the other quantities do not vanish.

One can construct a specific expression for the functions D^E_{lmn} in terms of the microscopic parameters of the medium by considering particular forms of the Hamiltonians.

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