ON THE ELECTRODYNAMICS OF WEAKLY NONLINEAR MEDIA

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Certain general properties of the second and third order electrical conductivity tensors, describing the nonlinear response of a medium to an external perturbation, are established.

1. INTRODUCTION

FOR the investigation of nonlinear effects in electrodynamics in those cases when these effects are small, it is natural to represent the current density j(t) (or the electric displacement vector **D**(t)) in the form of an expansion in powers of the macroscopic field **E**(t) and to confine one's attention to the linear and the first few nonlinear terms. The term of second order in the field differs from zero only in media without centers of symmetry (and also in media in which this symmetry is removed, for example, by the presence of a strong constant field). Owing to this, there is reason to consider also the third-order quantities along with the quantities of second order in the field. It turns out that one can represent the indicated expansion of the current density, correct to terms of the third order, in the form[1)]

$$\begin{aligned} j_i(t) = \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} e^{-i\omega t} \Big\{ & \sigma_{ij}^{(1)}(\omega) E_j(\omega) \\ & + \int_{-\infty}^{+\infty} \frac{d\omega_1}{2\pi} \sigma_{ijk}^{(2)}(\omega, \omega_1) E_j(\omega - \omega_1) E_k(\omega_1) \\ & + \int_{-\infty}^{+\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{+\infty} \frac{d\omega_2}{2\pi} \sigma_{ijkl}^{(3)}(\omega, \omega_1, \omega_2) E_j(\omega - \omega_1) \\ & \times E_k(\omega_1 - \omega_2) E_l(\omega_2) \Big\}. \end{aligned} \tag{1}$$

Here the $E_j(\omega)$ are the Fourier transforms of the field components $E_j(t)$; $\sigma_{ij}^{(1)}(\omega)$ is the complex electrical conductivity tensor, and one can call $\sigma_{ijk}^{(2)}(\omega, \omega_1)$ and $\sigma_{ijkl}^{(3)}(\omega, \omega_1, \omega_2)$ the electrical conductivity tensors of second and third orders, respectively. The expression for $D_i(t)$ differs formally from (1) only by replacing the electrical conductivity $\sigma^{(n)}$ by the dielectric constant $\epsilon^{(n)}$.

Recently, especially thanks to the development of laser technology, it has become possible to investigate the nonlinear properties of homogeneous media over an extremely wide frequency spectrum, including the optical band.[1] In this connection, an investigation of the general properties of the second and third order electrical conductivity tensors as functions of the frequency is of definite interest. In the present note, certain symmetry properties of these tensors and their analytic properties in complex frequency planes are established.

[1)]Spatial dispersion effects are not considered here.

2. GENERAL EXPRESSIONS FOR THE SECOND AND THIRD ORDER ELECTRICAL CONDUCTIVITY TENSORS

The Hamiltonian for the interaction of the system with the field **E**(t) is[2)]

$$H_E(t) = -P_i(t) E_i(t), \tag{2}$$

where **P**(t) is the dipole moment operator of the system.

A calculation based on perturbation theory for the Heisenberg current density operator (or for the density matrix), leads to the following expression for the observable current density:

$$\begin{aligned} j(t) = & \int_{-\infty}^{t} dt' \varphi_{ij}^{(1)}(t - t') E_j(t') \\ & + \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' \varphi_{ijk}^{(2)}(t - t', t' - t'') E_j(t') E_k(t'') \\ & + \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt'' \int_{-\infty}^{t''} dt''' \varphi_{ijkl}^{(3)}(t - t', t' - t'', t'' - t''') \\ & \times E_j(t') E_k(t'') E_l(t'''). \end{aligned} \tag{3}$$

The aftereffect functions are

$$\varphi_{ij}^{(1)}(\tau) = (i\hbar)^{-1} \langle [P_j, j_i(\tau)] \rangle, \tag{4a}$$

[2)]The question of whether the field **E**(t) in Eq. (2) is the same as the macroscopic field is not very clear (see [2]). This, however, is not too essential for our purposes.

$$\varphi^{(2)}_{ijk}(\tau, \tau_1) = (i\hbar)^{-2} \langle [P_k(-\tau_1), [P_j, j_i(\tau)]] \rangle, \quad (4b)$$

$$\varphi^{(3)}_{ijkl}(\tau, \tau_1, \tau_2) = (i\hbar)^{-3} \langle [P_l(-\tau_2 - \tau_1), [P_k(-\tau_1), [P_j, j_i(\tau)]]] \rangle, \quad (4c)$$

where the angle brackets denote the average with respect to the equilibrium ensemble. Expressions (3) and (4) for the current density and the aftereffect functions were derived in the articles by Kubo [3] and Bernard and Callen. [4]

We substitute the Fourier expansion of the field into Eq. (3). The expression for the current density takes the form (1), where (after symmetrization)

$$\sigma^{(1)}_{ij}(\omega) = \int_0^\infty d\tau e^{i\omega\tau} \varphi^{(1)}_{ij}(\tau), \quad (5a)$$

$$\sigma^{(2)}_{ijk}(\omega, \omega_1) = \frac{1}{2!}[\varphi^{(2)}_{ijk}(\omega, \omega_1) + \varphi^{(2)}_{ikj}(\omega, \omega - \omega_1)], \quad (5b)$$

$$\begin{aligned}\sigma^{(3)}_{ijkl}(\omega, \omega_1, \omega_2) = \frac{1}{3!}[&\varphi^{(3)}_{ijkl}(\omega, \omega_1, \omega_2) + \varphi^{(3)}_{ijlk}(\omega, \omega_1, \omega_1 - \omega_2) \\ &+ \varphi^{(3)}_{iljk}(\omega, \omega - \omega_2, \omega_1 - \omega_2) \\ &+ \varphi^{(3)}_{iklj}(\omega, \omega + \omega_2 - \omega_1, \omega - \omega_1) \\ &+ \varphi^{(3)}_{ikjl}(\omega, \omega - \omega_1 + \omega_2, \omega_2) \\ &+ \varphi^{(3)}_{ilkj}(\omega, \omega - \omega_2, \omega - \omega_1)], \end{aligned} \quad (5c)$$

where

$$\varphi^{(2)}_{ijk}(\omega, \omega_1) = \int_0^\infty d\tau \int_0^\infty d\tau_1 e^{i\omega\tau + i\omega_1\tau_1} \varphi^{(2)}_{ijk}(\tau, \tau_1), \quad (6a)$$

$$\varphi^{(3)}_{ijkl}(\omega, \omega_1, \omega_2) = \int_0^\infty d\tau \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 e^{i\omega\tau + i\omega_1\tau_1 + i\omega_2\tau_2} \varphi^{(3)}_{ijkl}(\tau, \tau_1, \tau_2). \quad (6b)$$

It is easy to verify that only the symmetric combinations (5b) and (5c), but not, for example, the tensors $\varphi^{(2)}_{ijk}(\omega, \omega_1)$ and $\varphi^{(2)}_{ikj}(\omega, \omega - \omega_1)$ separately, enter into the expression for j(t) (and consequently play a role in the experiments). The tensors $\sigma^{(2)}_{ijk}(\omega, \omega_1)$ and $\sigma^{(3)}_{ijkl}(\omega, \omega_1, \omega_2)$ satisfy the obvious symmetry relations (which we write only for $\sigma^{(2)}$):

$$\sigma^{(2)}_{ijk}(\omega, \omega_1) = \sigma^{(2)}_{ikj}(\omega, \omega - \omega_1). \quad (7)$$

The aftereffect functions must be real. This is not difficult to verify. It follows from the reality of the aftereffect functions that the complex conjugation operation on the electrical conductivity tensors is equivalent to changing the signs of all frequency variables. For example,

$$\sigma^{(3)*}_{ijkl}(\omega, \omega_1, \omega_2) = \sigma^{(3)}_{ijkl}(-\omega, -\omega_1, -\omega_2). \quad (8)$$

It follows from this that the real part of the electrical conductivity of arbitrary order is symmetric and the imaginary part antisymmetric with respect to simultaneously changing the signs of all frequency arguments.

3. ANALYTIC PROPERTIES AND DISPERSION RELATIONS

The integration in (6) is only carried out over positive times τ, τ_1, τ_2. This is, as is well known, none other than an expression of the principle of causality. As a consequence of the positiveness of τ, τ_1, and τ_2 the quantities $\varphi^{(2)}_{ijk}(\omega, \omega_1)$ and $\varphi^{(3)}_{ijkl}(\omega, \omega_1, \omega_2)$, regarded as functions of the complex frequencies ω, ω_1, and ω_2, are analytic functions of these variables in the corresponding upper half-planes (because of the presence of the exponential cut-off factors).

But the electrical conductivity tensors (5) (which are the only ones that are involved in experiments) are analytic functions in the entire upper half-plane only for the first frequency ω, i.e., the frequency at which the measurement is carried out! Actually, if the explicit expressions for $\sigma^{(2)}_{ijk}(\omega, \omega_1)$ and $\sigma^{(3)}_{ijkl}(\omega, \omega_1, \omega_2)$ are written out in accordance with (5) and (6), then it is easy to see that the variable ω enters into the exponential factors with the same sign (plus) in all terms.

It is also necessary to take it into account that as $\omega \to \infty$ all electrical conductivity tensors tend to zero; in addition, they do not have poles on the real axis. Hence dispersion relations of the Kramers-Kronig type exist between the real and imaginary parts of each of these tensors:

$$\operatorname{Re} \sigma^{(2)}_{ijk}(\omega, \omega_1) = \frac{1}{\pi} \mathrm{P} \int_{-\infty}^{+\infty} \frac{d\omega'}{\omega' - \omega} \operatorname{Im} \sigma^{(2)}_{ijk}(\omega', \omega_1),$$

$$\operatorname{Im} \sigma^{(2)}_{ijk}(\omega, \omega_1) = -\frac{1}{\pi} \mathrm{P} \int_{-\infty}^{+\infty} \frac{d\omega'}{\omega' - \omega} \operatorname{Re} \sigma^{(2)}_{ijk}(\omega', \omega_1) \quad (9)$$

and analogous relations for $\sigma^{(3)}_{ijk}(\omega, \omega_1, \omega_2)$.

Typical nonlinear effects involve electrical conductivity tensors with certain relations between the frequency variables. It is of interest to investigate their analytic properties. Thus, for example, it is easy to show that the effect of frequency doubling by means of a medium is determined by the tensor $\sigma^{(2)}_{ijk}(2\Omega, \Omega)$, and frequency tripling is determined by the tensor $\sigma^{(3)}_{ikjl}(3\Omega, 2\Omega, \Omega)$. It follows from (5) that these quantities are analytic functions of (only) the variable Ω in its upper half-plane. In writing down dispersion relations for these tensors, one can use (8) to transform the integrals over all frequencies into integrals over positive frequencies only.

The tensor $\sigma^{(2)}_{ijk}(\Omega_1 + \Omega_2, \Omega_1)$ determines the effect of the appearance of the sum frequency $\Omega_1 + \Omega_2$ upon superposition of two fields with frequencies Ω_1 and Ω_2, and the tensor $\sigma^{(3)}_{ijkl}(\Omega_1 + 2\Omega_2, 2\Omega_2, \Omega_2)$ determines the effect of the sum frequency $\Omega_1 + 2\Omega_2$. The elements of these tensors are analytic functions of both Ω_1 and Ω_2 in the upper half-planes of these variables.

The elements of the tensors $\sigma^{(2)}_{ijk}(\Omega_1 - \Omega_2, \Omega_1)$ and $\sigma^{(3)}_{ijkl}(2\Omega_1 - \Omega_2, 2\Omega_1, \Omega_1)$, which describe the effects of producing frequency differences ($\Omega_1 - \Omega_2$ and $2\Omega_1 - \Omega_2$, respectively), are analytic functions of Ω_1 in the upper half-plane and of Ω_2 in the lower half-plane.

The tensor $\sigma^{(3)}_{ijkl}(\Omega_1, 0, \Omega_2)$ describes the effect of radiation with frequency Ω_2 on the conductivity measured at the frequency Ω_1 (photoconductivity). The elements of this tensor are analytic functions of Ω_1 (in the upper half-plane), but are not analytic functions of Ω_2. The tensor $\sigma^{(2)}_{ijk}(0, \Omega)$, determining the effect of rectification of frequency Ω on a nonlinear medium, and the tensor $\sigma^{(3)}_{ijkl}(\Omega, 0, \Omega)$, determining the "self-action" of a field of frequency Ω, are not analytic in either the upper or lower half-planes of Ω.

In similar manner, it is possible to establish the analytic properties of the coefficients of reflection from a nonlinear medium (they exist in the form of reflection coefficients of second and third orders in the field of the incident wave).

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