Optical activity and spatial dispersion

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Constitutive equations for the description of optical activity are considered in the scheme of anisotropic nonconducting materials whose response is memory dependent and nonlocal. Attention is then restricted to models containing spatial derivatives up to second order. A dissipation principle is adopted in the form of the Clausius inequality for cycles and, because of nonlocality, the occurrence of an entropy flux is allowed. Thermodynamic restrictions are derived by accounting for the constraints placed by Maxwell's equations and letting the fields be time harmonic. Optically active (chiral) and optically inactive media are examined separately. In the first case thermodynamics is shown to imply the definiteness of the imaginary parts of the permittivity and the permeability along with a bound for the skew-symmetric terms of the real parts. In the second case the occurrence of quadratic terms or higher proves to rule out the possibility of first-order terms. [S1063-651X(97)08607-8]

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I. INTRODUCTION

Optical activity is generally described by letting the permittivity tensor be a function of the frequency ω (of monochromatic waves) and the wave vector $\mathbf{k}$. It is then understood that the pertinent fields are taken to depend on space and time as time-harmonic plane waves $\exp[i(\mathbf{k}\cdot\mathbf{x}-\omega t)]$. Next the dependence of the permittivity on $\mathbf{k}$ is expressed in terms of expansions in powers of $\mathbf{k}$ to the order of interest (see [1–3]). Hence, by considering suitable terms in $\mathbf{k}$ and disregarding others, various models are given (optically active and optically inactive media). It is the purpose of this paper to show that definite results are obtained by framing the behavior of electromagnetic media within the context of nonlocal materials, with memory, and by deriving the restrictions placed by thermodynamics.

In general, the value of the electric displacement at the place $\mathbf{x}$ and time t, $\mathbf{D}(\mathbf{x},t)$, depends on the value of the electric field $\mathbf{E}(\mathbf{x}',t')$ at all previous times $t'\leqslant t$ and places $\mathbf{x}'$ in some region about $\mathbf{x}$. We let the analogous property hold for the magnetic induction $\mathbf{B}$ and the magnetic field $\mathbf{H}$. Physical motivations for such nonlocal behavior, or spatial dispersion, are given, e.g., in [3,4]. Here we exhibit the essentials of a thermodynamic framework, for nonconducting materials, which incorporates nonlocality through an entropy flux. Next, starting from linear relations for materials with spatial dispersion and memory in time, approximate constitutive equations are considered that involve first- and second-order space derivatives.

The thermodynamic analysis is performed by preliminarily investigating the restrictions placed by Maxwell's equations. First, natural optical activity is considered through a general model that comprises chiral media. In particular it follows that the entropy flux is zero, the imaginary part of the permittivity tensor is positive (definite), the imaginary part of the inverse of the permeability tensor is negative, and their real parts have suitably small skew-symmetric terms. Simple models of the physical literature are shown to be recovered as particular cases. The interest in chiral media has been emphasized by recent publications, which have appeared in considerable abundance (see [5–7] and references therein). Next, optically inactive media are modeled by keeping all terms of spatial interaction up to second order in the distance. The entropy flux proves to be nonzero and the fourth-order tensors, relating the second-order derivatives of the electric field and the magnetic field to the electric displacement and the magnetic induction, satisfy relations analogous to those of the permittivity and the permeability.

II. DISSIPATION PRINCIPLE

Consider a body subject to an electromagnetic field and occupying a region $\mathcal{R}$. We assume that the material is nonconducting, i.e., the heat flux and the electric current vanish, and the (absolute) temperature θ is constant. Throughout, the rationalized system of units is used. Hence, on the basis of Poynting's theorem (see [8]), we write the balance of energy as

$$\dot{e}=\mathbf{H}\cdot\dot{\mathbf{B}}+\mathbf{E}\cdot\dot{\mathbf{D}}+r, \tag{2.1}$$

where e is the specific internal energy, r is the heat supply (per unit volume), and an overdot denotes the time derivative (see [9]). In standard approaches, an entropy density η is considered and the statement of the second law is taken to be in the Clausius-Duhem form

$$\dot{\eta}-\frac{r}{\theta}\geqslant 0 \tag{2.2}$$

for any process. Substitution of r from Eq. (2.1) into Eq. (2.2) gives

$$\theta\dot{\eta}-\dot{e}+\mathbf{H}\cdot\dot{\mathbf{B}}+\mathbf{E}\cdot\dot{\mathbf{D}}\geqslant 0. \tag{2.3}$$

Here we let the statement be more general than Eq. (2.3) in that we restrict attention to cycles (see [10]) and let an entropy flux $\mathbf{N}$ occur. Consider any region $\mathcal{P}\subset\mathcal{R}$ and any cycle in the time interval $[0,d)$. Hence we let the dissipation principle or the second law of thermodynamics be expressed by the Clausius-type inequality

$$\int_0^d \left[\int_{\mathcal{P}} (\theta \dot{\eta} - \dot{\varepsilon} + \mathbf{H}\cdot\dot{\mathbf{B}} + \mathbf{E}\cdot\dot{\mathbf{D}}) dv + \int_{\partial\mathcal{P}} \theta \mathbf{N}\cdot\mathbf{n}\, da \right] dt \geqslant 0, \tag{2.4}$$

where $\mathbf{n}$ is the unit outward normal. The overall contribution of $\mathbf{N}$ on $\mathcal{R}$ is required to be zero [see Eq. (3.7) of [4]] in that

$$\int_0^d \left[\int_{\partial\mathcal{R}} \theta \mathbf{N}\cdot\mathbf{n}\, da \right] dt = 0. \tag{2.5}$$

It is convenient to consider the free-enthalpy density $\zeta = e - \theta\eta - \mathbf{H}\cdot\mathbf{B} - \mathbf{E}\cdot\mathbf{D}$. In a cycle, at any place $\mathbf{x}$ of the body, the final value of ζ at time $t=d$ equals the initial value at time $t=0$. Upon substitution for ζ in Eq. (2.4) we have

$$\int_0^d \left[-\int_{\mathcal{P}} (\dot{\mathbf{H}}\cdot\mathbf{B} + \dot{\mathbf{E}}\cdot\mathbf{D}) dv + \int_{\partial\mathcal{P}} \theta\mathbf{N}\cdot\mathbf{n}\, da \right] dt \geqslant 0. \tag{2.6}$$

For the present purposes the inequality (2.6) is allowed to hold for any smooth region $\mathcal{P}$. Hence the smoothness of the fields, the divergence theorem, and the arbitrariness of $\mathcal{P}$ yield

$$\int_0^d [\dot{\mathbf{H}}\cdot\mathbf{B} + \dot{\mathbf{E}}\cdot\mathbf{D} - \nabla\cdot(\theta\mathbf{N})](\mathbf{x},t)\, dt \leqslant 0 \tag{2.7}$$

for every place $\mathbf{x}$ of the body.

Any physically admissible model of electromagnetic (nonconducting) solid must satisfy the inequality (2.7) with an appropriate expression of the entropy flux $\mathbf{N}$. Indeed, as we will see in a moment, the form of $\mathbf{N}$ is strictly related to the constitutive model under consideration.

III. CONSTITUTIVE RELATIONS

To describe optical activity we may borrow from the scheme of materials with memory where the state is a proper set of histories [9,10]. Indeed, we let the state of the material be the pair of fields of histories $\mathbf{E}^t, \mathbf{H}^t$ at the pertinent region $\mathcal{P}$. Hence we write

$$\mathbf{D}(\mathbf{x},t) = \tilde{\mathbf{D}}(\mathbf{E}^t(\ ),\mathbf{x}), \quad \mathbf{B}(\mathbf{x},t) = \tilde{\mathbf{B}}(\mathbf{H}^t(\ ),\mathbf{x}), \tag{3.1}$$

with the meaning that, e.g., $\mathbf{D}$ at the place $\mathbf{x}$ and time t depends on the electric field $\mathbf{E}$ at any place at all times prior to the present time t. Denote by $\mathcal{P}_{\mathbf{x}}$ the region $\mathcal{P}$ relative to the origin $\mathbf{x}$. Here the functionals $\tilde{\mathbf{D}}, \tilde{\mathbf{B}}$ are taken to be linear in that

$$\mathbf{D}(\mathbf{x},t) = \int_{\mathcal{P}_{\mathbf{x}}} \left[\boldsymbol{\tau}(\mathbf{r})\mathbf{E}(\mathbf{x}+\mathbf{r},t) + \int_0^\infty \boldsymbol{\chi}(\mathbf{r},u)\mathbf{E}(\mathbf{x}+\mathbf{r},t-u)\, du \right] dv, \tag{3.2}$$

and analogously for $\mathbf{B}$; $\boldsymbol{\tau}$ and $\boldsymbol{\chi}$ are functions that associate $\mathbf{r}\in\mathcal{P}_{\mathbf{x}}$, and $\mathbf{r}\in\mathcal{P}_{\mathbf{x}}, u\in[0,\infty)$ with a second-order tensor. For smooth fields we write

$$\mathbf{E}(\mathbf{x}+\mathbf{r},t-u) = \mathbf{E}(\mathbf{x},t-u) + (\mathbf{r}\cdot\nabla)\mathbf{E}(\mathbf{x},t-u) + \tfrac{1}{2}(\mathbf{r}\otimes\mathbf{r})\cdot(\nabla\otimes\nabla)\mathbf{E}(\mathbf{x},t-u) + o(\mathbf{r}^2), \tag{3.3}$$

where $o(\mathbf{r}^2)\to 0$ faster than $\mathbf{r}^2$, uniformly in $t-u$. Substitution of Eq. (3.3) into Eq. (3.2) gives

$$D_i(t) = \varepsilon_{ij} * E_j^t + \beta_{ijk} * E_{[j,k]}^t + \beta_{ijk} * E_{(j,k)} + \gamma_{ijkl} * E_{j,kl}^t + \text{h.o.}, \tag{3.4}$$

where the asterisk denotes a generalized convolution, e.g.,

$$\varepsilon_{ij} * E_j^t = \varepsilon_{ij}^0 E_j(t) + \int_0^\infty \varepsilon_{ij}(u) E_j(t-u)\, du. \tag{3.5}$$

The constant terms $\varepsilon_{ij}^0, \beta_{ijk}^0, \gamma_{ijkl}^0$ and the functions $\varepsilon_{ij}(u), \beta_{ijk}(u), \gamma_{ijkl}(u)$ are the integrals over $\mathcal{P}_{\mathbf{x}}$ of $\tau_{ij}(\mathbf{r}), \tau_{ij}(\mathbf{r})r_k, \tau_{ij}, (\mathbf{r})r_k r_l$ and $\chi_{ij}(\mathbf{r},u), \chi_{ij}(\mathbf{r},u)r_k$, $\chi_{ij}(\mathbf{r},u)r_k r_l$. Also, h.o. means the remaining contribution due to the integrals of $\boldsymbol{\tau}(\mathbf{r})o(\mathbf{r}^2)$, and $\boldsymbol{\chi}(\mathbf{r},u)o(\mathbf{r}^2)$, while (k,j) and $[k,j]$ denote symmetrization and skew symmetrization. The skew part $[k,j]$ allows the second term to be written as the ith component of, say, $\boldsymbol{\alpha}\nabla\times\mathbf{E}$, where $\alpha_{is} = \frac{1}{2}\beta_{ijk}\varepsilon_{sjk}$ and ε_{sjk} is the sjk component of the alternating tensor. The same arguments apply to the expression for $\mathbf{B}$.

Henceforth we disregard the h.o. terms. Hence we write the constitutive equations (3.1) for $\mathbf{D}$ and $\mathbf{B}$ as

$$D_i(t) = \varepsilon_{ij} * E_k^t + \alpha_{ij} * (\nabla\times\mathbf{E}^t)_j + \beta_{ijk} * E_{(j,k)}^t + \gamma_{ijkl} * E_{j,kl}^t, \tag{3.6}$$

$$B_i(t) = \mu_{ij} * H_j^t + \lambda_{ij} * (\nabla\times\mathbf{H}^t)_j + \nu_{ijk} * H_{(j,k)}^t + \kappa_{ijkl} * H_{j,kl}^t. \tag{3.7}$$

The state of the body, at the place $\mathbf{x}$ and time t, is then the set of histories $\mathbf{E}^t(\mathbf{x}), \nabla\mathbf{E}^t(\mathbf{x}), \nabla\nabla\mathbf{E}^t(\mathbf{x}), \mathbf{H}^t(\mathbf{x}), \nabla\mathbf{H}^t(\mathbf{x})$, $\nabla\nabla\mathbf{H}^t(\mathbf{x})$. Incidentally, the knowledge of the field $\mathbf{E}^t(\ )$ allows the evaluation of the gradients $\nabla\mathbf{E}^t(\ ), \nabla\nabla\mathbf{E}^t(\ )$ at any place. Here, however, the state involves the history at the pertinent place $\mathbf{x}$, not the whole field $\mathbf{E}^t(\ )$. Hence $\mathbf{E}^t(\mathbf{x}), \nabla\mathbf{E}^t(\mathbf{x}), \nabla\nabla\mathbf{E}^t(\mathbf{x})$ are independent of one another.

If the memory effects are negligible, Eqs. (3.6) and (3.7) simplify to the corresponding ones with the convolutions replaced by constant coefficients times the value at time t. Substitution of Eqs. (3.6) and (3.7) into the inequality (2.7) and some rearrangement yield

$$\int_0^d [-(\dot{\mathbf{H}}\cdot\boldsymbol{\mu} * \mathbf{H} + \dot{\mathbf{H}}\cdot\boldsymbol{\lambda} * \nabla\times\mathbf{H} + \dot{\mathbf{H}}\cdot\boldsymbol{\nu} * \nabla\mathbf{H} - \nabla\dot{\mathbf{H}}\cdot\boldsymbol{\kappa} * \nabla\mathbf{H} + \dot{\mathbf{E}}\cdot\boldsymbol{\varepsilon} * \mathbf{E} + \dot{\mathbf{E}}\cdot\boldsymbol{\alpha} * \nabla\times\mathbf{E} + \dot{\mathbf{E}}\cdot\boldsymbol{\beta} * \nabla\mathbf{E} - \nabla\dot{\mathbf{E}}\cdot\boldsymbol{\gamma} * \nabla\mathbf{E}) + \nabla\cdot(\theta\mathbf{N} - \dot{\mathbf{E}}\boldsymbol{\gamma} * \nabla\mathbf{E} - \dot{\mathbf{H}}\boldsymbol{\kappa} * \nabla\mathbf{H})]\, dt \geqslant 0, \tag{3.8}$$

where, e.g., $\dot{\mathbf{H}}\cdot\boldsymbol{\nu} * \nabla\mathbf{H} = \dot{\mathbf{H}}_i \nu_{ijk} * H_{j,k}$. Hence Eq. (3.8) holds only if

$$\theta\mathbf{N} = \dot{\mathbf{E}}\boldsymbol{\gamma} * \nabla\mathbf{E} + \dot{\mathbf{H}}\boldsymbol{\kappa} * \nabla\mathbf{H} \tag{3.9}$$

to within the curl of any vector function. We now examine the remaining inequality

$$\int_0^d [-(\dot{\mathbf{H}}\cdot\boldsymbol{\mu}*\mathbf{H}+\dot{\mathbf{H}}\cdot\boldsymbol{\lambda}*\nabla\times\mathbf{H}+\dot{\mathbf{H}}\cdot\boldsymbol{\nu}*\nabla\mathbf{H}-\nabla\dot{\mathbf{H}}\cdot\boldsymbol{\kappa}*\nabla\mathbf{H} +\dot{\mathbf{E}}\cdot\boldsymbol{\varepsilon}*\mathbf{E}+\dot{\mathbf{E}}\cdot\boldsymbol{\alpha}*\nabla\times\mathbf{E}+\dot{\mathbf{E}}\cdot\boldsymbol{\beta}*\nabla\mathbf{E} -\nabla\dot{\mathbf{E}}\cdot\boldsymbol{\gamma}*\nabla\mathbf{E})]dt\geqslant 0, \tag{3.10}$$

which must hold for all admissible fields at any place $\mathbf{x}$ in $\mathcal{R}$. To derive the restrictions placed by Eq. (3.10) on the constitutive equations (3.6) and (3.7) it is convenient to consider time-harmonic functions. If the functions $\mathbf{E}$ and $\mathbf{H}$ are time harmonic, i.e.,

$$\mathbf{E}(\mathbf{x},t)=\boldsymbol{\mathcal{E}}(\mathbf{x})\exp(-i\omega t),\quad \mathbf{H}(\mathbf{x},t)=\boldsymbol{\mathcal{H}}(\mathbf{x})\exp(-i\omega t), \tag{3.11}$$

the time-dependence factorizes, i.e.,

$$\mathbf{D}(\mathbf{x},t)=\boldsymbol{\mathcal{D}}(\mathbf{x})\exp(-i\omega t),\quad \mathbf{B}(\mathbf{x},t)=\boldsymbol{\mathcal{B}}(\mathbf{x})\exp(-i\omega t), \tag{3.12}$$

and the constitutive equations become

$$\mathcal{D}_i=\hat{\varepsilon}_{ij}\mathcal{E}_j+\hat{\alpha}_{ij}(\nabla\times\boldsymbol{\mathcal{E}})_j+\hat{\beta}_{ijk}\mathcal{E}_{(j,k)}+\hat{\gamma}_{ijkl}\mathcal{E}_{j,kl}, \tag{3.13}$$

$$\mathcal{B}_i=\hat{\mu}_{ij}\mathcal{H}_j+\hat{\lambda}_{ij}(\nabla\times\boldsymbol{\mathcal{H}})_j+\hat{\nu}_{ijk}\mathcal{H}_{(j,k)}+\hat{\kappa}_{ijkl}\mathcal{H}_{j,kl}, \tag{3.14}$$

where the caret denotes the coefficient of the instantaneous response plus the Fourier transform of the kernel, e.g.,

$$\hat{\varepsilon}_{ij}=\varepsilon^0_{ij}+\int_0^\infty \varepsilon_{ij}(u)\exp(i\omega u)du =\varepsilon^0_{ij}+\int_0^\infty \varepsilon_{ij}(u)\cos\omega u\; du+i\int_0^\infty \varepsilon_{ij}(u)\sin\omega u\; du. \tag{3.15}$$

If the material is heterogeneous the coefficients $\hat{\varepsilon}_{ij},\ldots,\hat{\kappa}_{ijkl}$ depend on the place $\mathbf{x}$; it is understood that $\boldsymbol{\mathcal{D}},\boldsymbol{\mathcal{B}},\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{H}}$ and the coefficients $\hat{\varepsilon}_{ij},\ldots,\hat{\kappa}_{ijkl}$ are evaluated at the same place. Since a cycle has to occur from $t=0$ to d, by Eqs. (3.11) and (3.12) we set $d=2\pi/\omega$.

The physical content of Eq. (3.10) is preserved provided we replace each term with the real part of the appropriate time-harmonic function. In this regard consider the identity

$$\mathrm{Re}[\mathbf{v}\exp(-i\omega t)]\cdot\mathrm{Re}[\mathbf{w}\exp(-i\omega t)] =\tfrac{1}{4}[\mathbf{v}\cdot\mathbf{w}^*+\mathbf{v}^*\cdot\mathbf{w}+\mathbf{v}\cdot\mathbf{w}\exp(-2i\omega t) +\mathbf{v}^*\cdot\mathbf{w}^*\exp(2i\omega t)], \tag{3.16}$$

where the asterisk superscript means a complex conjugate, and observe that the contributions of $\exp(-2i\omega t)$ and $\exp(2i\omega t)$ vanish upon integration on the period $[0,2\pi/\omega)$. Hence we obtain from Eq. (3.10) that

$$i\boldsymbol{\mathcal{H}}\cdot(\hat{\boldsymbol{\mu}}\boldsymbol{\mathcal{H}})^*+i\boldsymbol{\mathcal{H}}\cdot(\hat{\boldsymbol{\lambda}}\nabla\times\boldsymbol{\mathcal{H}})^*+i\boldsymbol{\mathcal{H}}\cdot(\hat{\boldsymbol{\nu}}\nabla\boldsymbol{\mathcal{H}})^* -i\nabla\boldsymbol{\mathcal{H}}\cdot(\boldsymbol{\kappa}\nabla\boldsymbol{\mathcal{H}})^*+i\boldsymbol{\mathcal{E}}\cdot(\hat{\boldsymbol{\varepsilon}}\boldsymbol{\mathcal{E}})^*+i\boldsymbol{\mathcal{E}}\cdot(\hat{\boldsymbol{\alpha}}\nabla\times\boldsymbol{\mathcal{E}})^* +i\boldsymbol{\mathcal{E}}\cdot(\hat{\boldsymbol{\beta}}\nabla\boldsymbol{\mathcal{E}})^*-i\nabla\boldsymbol{\mathcal{E}}\cdot(\boldsymbol{\gamma}\nabla\boldsymbol{\mathcal{E}})^*+\text{c.c.}\geqslant 0, \tag{3.17}$$

where c.c. denotes the complex conjugate of the whole preceding part. The fields $\boldsymbol{\mathcal{E}}$ and $\boldsymbol{\mathcal{H}}$ are required to satisfy Maxwell's equations. For time-harmonic fields, Faraday's law and Ampère's law reduce to

$$\nabla\times\boldsymbol{\mathcal{E}}=i\omega\boldsymbol{\mathcal{B}},\quad \nabla\times\boldsymbol{\mathcal{H}}=-i\omega\boldsymbol{\mathcal{D}}+\boldsymbol{\mathcal{J}}, \tag{3.18}$$

where $\boldsymbol{\mathcal{J}}$ is the phasor of the current density $\mathbf{J}$, i.e., $\mathbf{J}(\mathbf{x},t)=\boldsymbol{\mathcal{J}}(\mathbf{x})\exp(-i\omega t)$. Also, setting $\rho(\mathbf{x},t)=\varrho(\mathbf{x})\exp(-i\omega t)$ be the free-charge density, we write the continuity equation as

$$\nabla\cdot\boldsymbol{\mathcal{J}}=i\omega\varrho. \tag{3.19}$$

It follows from Eqs. (3.18) and (3.19) that

$$\nabla\cdot\boldsymbol{\mathcal{B}}=0,\quad \nabla\cdot\boldsymbol{\mathcal{D}}=\varrho. \tag{3.20}$$

Hence the fields $\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{H}},\boldsymbol{\mathcal{B}},\boldsymbol{\mathcal{D}}$ satisfy Maxwell's equations provided only that Eqs. (3.18) and (3.19) hold.

IV. ANISOTROPIC CHIRAL MEDIA

A remarkable particular case is obtained by setting $\hat{\boldsymbol{\beta}},\hat{\boldsymbol{\gamma}},\hat{\boldsymbol{\nu}},\hat{\boldsymbol{\kappa}}=0$, whence

$$\boldsymbol{\mathcal{D}}=\hat{\boldsymbol{\varepsilon}}\boldsymbol{\mathcal{E}}+\hat{\boldsymbol{\alpha}}\nabla\times\boldsymbol{\mathcal{E}},\quad \boldsymbol{\mathcal{B}}=\hat{\boldsymbol{\mu}}\boldsymbol{\mathcal{H}}+\hat{\lambda}\nabla\times\boldsymbol{\mathcal{H}}. \tag{4.1}$$

Hence Eq. (3.9) implies that

$$\mathbf{N}=\mathbf{0}. \tag{4.2}$$

Owing to Eq. (3.18) we have

$$\boldsymbol{\mathcal{D}}=\hat{\boldsymbol{\varepsilon}}\boldsymbol{\mathcal{E}}+i\omega\hat{\boldsymbol{\alpha}}\boldsymbol{\mathcal{B}} \tag{4.3}$$

and

$$\boldsymbol{\mathcal{B}}=\hat{\boldsymbol{\mu}}\boldsymbol{\mathcal{H}}-i\omega\hat{\lambda}\hat{\boldsymbol{\varepsilon}}\boldsymbol{\mathcal{E}}+\omega^2\hat{\boldsymbol{\lambda}}\hat{\boldsymbol{\alpha}}\boldsymbol{\mathcal{B}}. \tag{4.4}$$

If $\hat{\boldsymbol{\mu}}$ is nonsingular Eq. (4.4) gives

$$\boldsymbol{\mathcal{H}}=i\omega\hat{\boldsymbol{\xi}}\boldsymbol{\mathcal{E}}+\hat{\boldsymbol{\eta}}\boldsymbol{\mathcal{B}}, \tag{4.5}$$

where

$$\hat{\boldsymbol{\xi}}=\hat{\boldsymbol{\mu}}^{-1}\hat{\boldsymbol{\lambda}}\hat{\boldsymbol{\varepsilon}},\quad \hat{\boldsymbol{\eta}}=\hat{\boldsymbol{\mu}}^{-1}(1-\omega^2\hat{\boldsymbol{\lambda}}\hat{\boldsymbol{\alpha}}). \tag{4.6}$$

Accordingly, for any pair of (complex) values $\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{B}}$, the admissible values of $\boldsymbol{\mathcal{D}}$ and $\boldsymbol{\mathcal{H}}$ are given by Eqs. (4.3) and (4.5) and those of $\nabla\times\boldsymbol{\mathcal{E}}$ and $\nabla\times\boldsymbol{\mathcal{H}}$ by Eq. (3.18). The inequality (3.17) reduces to

$$i(i\omega\hat{\boldsymbol{\xi}}\boldsymbol{\mathcal{E}}+\hat{\boldsymbol{\eta}}\boldsymbol{\mathcal{B}})\cdot\boldsymbol{\mathcal{B}}^*+i\boldsymbol{\mathcal{E}}\cdot(\hat{\boldsymbol{\varepsilon}}\boldsymbol{\mathcal{E}}^*-i\omega\hat{\boldsymbol{\alpha}}^*\boldsymbol{\mathcal{B}}^*)+\text{c.c.}\geqslant 0, \tag{4.7}$$

which must hold for arbitrary values of $\boldsymbol{\mathcal{E}}$ and $\boldsymbol{\mathcal{B}}$. Let the subscripts 1 and 2 indicate the real and imaginary parts of a quantity, e.g., $\hat{\boldsymbol{\varepsilon}}=\hat{\boldsymbol{\varepsilon}}_1+i\hat{\boldsymbol{\varepsilon}}_2$ and $\boldsymbol{\mathcal{E}}=\boldsymbol{\mathcal{E}}_1+i\boldsymbol{\mathcal{E}}_2$. Letting a superscript T denote the transpose, we find from Eq. (4.7) that the inequality

$$-\boldsymbol{\mathcal{B}}_1\cdot\hat{\boldsymbol{\eta}}_2\boldsymbol{\mathcal{B}}_1-\boldsymbol{\mathcal{B}}_2\cdot\hat{\boldsymbol{\eta}}_2\boldsymbol{\mathcal{B}}_2+\boldsymbol{\mathcal{B}}_2\cdot(\hat{\boldsymbol{\eta}}_1-\hat{\boldsymbol{\eta}}_1^T)\boldsymbol{\mathcal{B}}_1+\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\varepsilon}}_2\boldsymbol{\mathcal{E}}_1$$
$$+\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\varepsilon}}_2\boldsymbol{\mathcal{E}}_2+\boldsymbol{\mathcal{E}}_1\cdot(\hat{\boldsymbol{\varepsilon}}_1-\hat{\boldsymbol{\varepsilon}}_1^T)\boldsymbol{\mathcal{E}}_2-\boldsymbol{\mathcal{E}}_1\cdot(\hat{\boldsymbol{\alpha}}_2+\hat{\boldsymbol{\xi}}_2^T)\boldsymbol{\mathcal{B}}_2$$
$$+\boldsymbol{\mathcal{E}}_2\cdot(\hat{\boldsymbol{\alpha}}_2+\hat{\boldsymbol{\xi}}_2^T)\boldsymbol{\mathcal{B}}_1+\boldsymbol{\mathcal{E}}_1\cdot(\hat{\boldsymbol{\alpha}}_1-\hat{\boldsymbol{\xi}}_1^T)\boldsymbol{\mathcal{B}}_1$$
$$+\boldsymbol{\mathcal{E}}_2\cdot(\hat{\boldsymbol{\alpha}}_1-\hat{\boldsymbol{\xi}}_1^T)\boldsymbol{\mathcal{B}}_2\geqslant 0 \qquad (4.8)$$

must hold for every quadruple of real vectors $\boldsymbol{\mathcal{E}}_1,\boldsymbol{\mathcal{E}}_2,\boldsymbol{\mathcal{B}}_1,\boldsymbol{\mathcal{B}}_2$. Hence it follows that

$$\hat{\boldsymbol{\eta}}_2<0,\quad \hat{\boldsymbol{\varepsilon}}_2>0,\quad \hat{\boldsymbol{\alpha}}_2=-\hat{\boldsymbol{\xi}}_2^T,\quad \hat{\boldsymbol{\alpha}}_1=\hat{\boldsymbol{\xi}}_1^T, \qquad (4.9)$$

while $\hat{\boldsymbol{\varepsilon}}_1-\hat{\boldsymbol{\varepsilon}}_1^T$ and $\hat{\boldsymbol{\eta}}_1-\hat{\boldsymbol{\eta}}_1^T$ are bounded by

$$-\boldsymbol{\mathcal{B}}_1\cdot\hat{\boldsymbol{\eta}}_2\boldsymbol{\mathcal{B}}_1-\boldsymbol{\mathcal{B}}_2\cdot\hat{\boldsymbol{\eta}}_2\boldsymbol{\mathcal{B}}_2+\boldsymbol{\mathcal{B}}_2\cdot(\hat{\boldsymbol{\eta}}_1-\hat{\boldsymbol{\eta}}_1^T)\boldsymbol{\mathcal{B}}_1\geqslant 0, \qquad (4.10)$$

$$\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\varepsilon}}_2\boldsymbol{\mathcal{E}}_1+\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\varepsilon}}_2\boldsymbol{\mathcal{E}}_2+\boldsymbol{\mathcal{E}}_1\cdot(\hat{\boldsymbol{\varepsilon}}_1-\hat{\boldsymbol{\varepsilon}}_1^T)\boldsymbol{\mathcal{E}}_2\geqslant 0. \qquad (4.11)$$

The symmetry relations $\hat{\boldsymbol{\varepsilon}}_1=\hat{\boldsymbol{\varepsilon}}_1^T$ and $\hat{\boldsymbol{\eta}}_1=\hat{\boldsymbol{\eta}}_1^T$, together with Eq. (4.9), are sufficient but not necessary for the inequalities (4.9) and (4.10) to hold. If the medium is isotropic the tensors $\hat{\boldsymbol{\varepsilon}},\hat{\boldsymbol{\alpha}},\hat{\boldsymbol{\mu}},\hat{\boldsymbol{\lambda}}$ become scalars $\hat{\varepsilon},\hat{\alpha},\hat{\mu},\hat{\lambda}$ times the identity tensor **1** and the whole set of conditions (4.9) reduces to

$$[(1-\omega^2\hat{\lambda}\hat{\alpha})/\hat{\mu}]_2<0,\quad \hat{\varepsilon}_2>0,$$
$$\hat{\alpha}_2=-(\hat{\lambda}\hat{\varepsilon}/\hat{\mu})_2,\quad \hat{\alpha}_1=(\hat{\lambda}\hat{\varepsilon}/\hat{\mu})_1. \qquad (4.12)$$

A further simplification arises if the medium is taken to be nondissipative. Formally, we specify the nondissipative character by requiring that Eq. (4.8) hold as an equality. The inequalities in Eq. (4.12) then become

$$[(1-\omega^2\hat{\lambda}\hat{\alpha})/\hat{\mu}]_2=0,\quad \hat{\varepsilon}_2=0. \qquad (4.13)$$

Now $\hat{\varepsilon}$ is real and the equalities in Eq. (4.12) amount to $\hat{\varepsilon}\hat{\lambda}/\hat{\mu}=\hat{\alpha}^*$. Substitution in Eq. (4.13) shows that also $\hat{\mu}$ is real; we then write ε and μ in place of $\hat{\varepsilon}$ and $\hat{\mu}$ to emphasize that they are real valued. Accordingly, we have

$$\hat{\alpha}=\frac{\varepsilon}{\mu}\hat{\lambda}^*. \qquad (4.14)$$

Back to the form (4.1) of the constitutive equations, by Eq. (4.14) we can write

$$\boldsymbol{\mathcal{D}}=\varepsilon\left[\boldsymbol{\mathcal{E}}+\frac{\hat{\lambda}^*}{\mu}\nabla\times\boldsymbol{\mathcal{E}}\right], \qquad (4.15)$$

$$\boldsymbol{\mathcal{B}}=\mu\left[\boldsymbol{\mathcal{H}}+\frac{\hat{\lambda}}{\mu}\nabla\times\boldsymbol{\mathcal{H}}\right]. \qquad (4.16)$$

If we let $\hat{\lambda}$ be real, Eqs. (4.15) and (4.16) are exactly the model that traces back to Drude, Born, and Federov (see [5]).

V. SECOND-ORDER SPATIAL DISPERSION

We consider the constitutive relations (3.13) and (3.14), which involve also second-order spatial derivatives, and investigate the restrictions placed by the inequality (3.17) on the coefficients $\hat{\boldsymbol{\varepsilon}},\hat{\boldsymbol{\alpha}},\ldots,\hat{\boldsymbol{\kappa}}$. In this regard we have to ascertain the degree of arbitrariness of the phasors $\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{H}},\boldsymbol{\mathcal{D}},\boldsymbol{\mathcal{B}}$ and their spatial derivatives. If the values of $\nabla\times\boldsymbol{\mathcal{E}}$ and $\nabla\times\boldsymbol{\mathcal{H}}$ are chosen arbitrarily, Eqs. (3.18) determine the values of $\boldsymbol{\mathcal{B}}$ and $\boldsymbol{\mathcal{D}}$. Hence Eqs. (3.13) and (3.14) hold with arbitrary values of $\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{H}}$ and of the symmetric derivatives $\mathcal{E}_{(i,j)},\mathcal{H}_{(i,j)}$ provided only that appropriate values of the second-order derivatives $\nabla\nabla\boldsymbol{\mathcal{E}}$ and $\nabla\nabla\boldsymbol{\mathcal{H}}$ can be found (nondegenerate case). Accordingly, at any point of the body the values of $\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{H}},\nabla\boldsymbol{\mathcal{E}},\nabla\boldsymbol{\mathcal{H}}$ can be regarded as arbitrary. This implies that the inequality (3.17) must hold for arbitrary values of $\boldsymbol{\mathcal{E}},\boldsymbol{\mathcal{H}},\nabla\boldsymbol{\mathcal{E}},\nabla\boldsymbol{\mathcal{H}}$.

By still using the subscripts 1 and 2 for the real and imaginary parts, it follows from Eq. (3.17) that

$$\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\varepsilon}}_2\boldsymbol{\mathcal{E}}_1+\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\varepsilon}}_2\boldsymbol{\mathcal{E}}_2+\boldsymbol{\mathcal{E}}_1\cdot(\hat{\boldsymbol{\varepsilon}}_1-\hat{\boldsymbol{\varepsilon}}_1^T)\boldsymbol{\mathcal{E}}_2+\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\alpha}}_2\nabla\times\boldsymbol{\mathcal{E}}_2$$
$$+\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\alpha}}_2\nabla\times\boldsymbol{\mathcal{E}}_1+\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\alpha}}_1\nabla\times\boldsymbol{\mathcal{E}}_2-\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\alpha}}_1\nabla\times\boldsymbol{\mathcal{E}}_1$$
$$+\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\beta}}_2\nabla\boldsymbol{\mathcal{E}}_1+\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\beta}}_2\nabla\boldsymbol{\mathcal{E}}_2+\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\beta}}_1\nabla\boldsymbol{\mathcal{E}}_2$$
$$-\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\beta}}_1\nabla\boldsymbol{\mathcal{E}}_1-\nabla\boldsymbol{\mathcal{E}}_1\cdot\hat{\boldsymbol{\gamma}}_2\nabla\boldsymbol{\mathcal{E}}_1-\nabla\boldsymbol{\mathcal{E}}_2\cdot\hat{\boldsymbol{\gamma}}_2\nabla\boldsymbol{\mathcal{E}}_2$$
$$+\nabla\boldsymbol{\mathcal{E}}_1\cdot(\hat{\boldsymbol{\gamma}}_1-\hat{\boldsymbol{\gamma}}_1^T)\nabla\boldsymbol{\mathcal{E}}_2+(\boldsymbol{\mathcal{E}}\mapsto\boldsymbol{\mathcal{H}})\geqslant 0, \qquad (5.1)$$

where $(\boldsymbol{\mathcal{E}}\mapsto\boldsymbol{\mathcal{H}})$ represents the analogous terms obtained by letting $\boldsymbol{\mathcal{E}}\mapsto\boldsymbol{\mathcal{H}}$ and changing appropriately the tensors involved. The arbitrariness of $\boldsymbol{\mathcal{E}}_1,\boldsymbol{\mathcal{E}}_2,\nabla\boldsymbol{\mathcal{E}}_1,\nabla\boldsymbol{\mathcal{E}}_2$ and $\boldsymbol{\mathcal{H}}_1,\boldsymbol{\mathcal{H}}_2,\nabla\boldsymbol{\mathcal{H}}_1,\nabla\boldsymbol{\mathcal{H}}_2$ allows us to conclude that Eq. (5.1) holds if and only if

$$\hat{\boldsymbol{\alpha}},\hat{\boldsymbol{\beta}},\hat{\boldsymbol{\lambda}},\hat{\boldsymbol{\nu}}=\mathbf{0}, \qquad (5.2)$$

$$\hat{\boldsymbol{\varepsilon}}_1:\ \mathbf{u}\cdot\hat{\boldsymbol{\varepsilon}}_2\mathbf{u}+\mathbf{v}\cdot\hat{\boldsymbol{\varepsilon}}_2\mathbf{v}+\mathbf{u}\cdot(\hat{\boldsymbol{\varepsilon}}_1-\hat{\boldsymbol{\varepsilon}}_1^T)\mathbf{v}\geqslant 0,\ \ \hat{\boldsymbol{\varepsilon}}_2>\mathbf{0} \qquad (5.3)$$

for all vectors **u**,**v**, and analogously for $\hat{\boldsymbol{\mu}}_2,\hat{\boldsymbol{\mu}}_1$, while

$$\hat{\boldsymbol{\gamma}}_1:\ \mathbf{A}\cdot\hat{\boldsymbol{\gamma}}_2\mathbf{A}+\mathbf{C}\cdot\hat{\boldsymbol{\gamma}}_2\mathbf{C}-\mathbf{A}\cdot(\hat{\boldsymbol{\gamma}}_1-\hat{\boldsymbol{\gamma}}_1^T)\mathbf{C}\leqslant 0,\quad \hat{\boldsymbol{\gamma}}_2<\mathbf{0} \qquad (5.4)$$

for all tensors **A**,**C**, and analogously for $\hat{\boldsymbol{\kappa}}_2,\hat{\boldsymbol{\kappa}}_1$.

Nondissipative media are again characterized by requiring that the dissipation inequality, here Eq. (5.1), hold as an equality. Such is the case if and only if, in addition, $\hat{\boldsymbol{\varepsilon}}_2,\hat{\boldsymbol{\mu}}_2,\hat{\boldsymbol{\gamma}}_2,\hat{\boldsymbol{\kappa}}_2=\mathbf{0}$ and, by omitting the unnecessary subscript 1,

$$\hat{\boldsymbol{\varepsilon}}=\hat{\boldsymbol{\varepsilon}}^T,\quad \hat{\boldsymbol{\mu}}=\hat{\boldsymbol{\mu}}^T,\quad \hat{\boldsymbol{\gamma}}=\hat{\boldsymbol{\gamma}}^T,\quad \hat{\boldsymbol{\kappa}}=\hat{\boldsymbol{\kappa}}^T. \qquad (5.5)$$

In indicial form, the symmetry for $\hat{\boldsymbol{\gamma}}$ and $\hat{\boldsymbol{\kappa}}$ in Eq. (5.5) means that

$$\hat{\gamma}_{ijkl}=\hat{\gamma}_{jikl},\quad \hat{\kappa}_{ijkl}=\hat{\kappa}_{jikl}, \qquad (5.6)$$

the invariance by interchange of the third and fourth indices being true by definition.

VI. CONCLUSION

Chiral media are considered by letting **D** be given by convolutions of **E** and $\nabla\times$**E** and **B** by convolutions of **H** and $\nabla\times$**H**. The constraints placed by Maxwell's equations are examined and hence necessary and sufficient conditions on the constitutive functions are derived for the second law to hold. If instead terms up to second-order derivatives are involved, the second law implies that the coefficients of the first-order derivatives vanish. A generalization of the model with higher-order derivatives would lead to the vanishing of the odd-order derivatives. This result allows us to view optically active or inactive media in a different way. If terms of second order and higher are zero, the medium can be optically active with appropriate thermodynamic restrictions. If higher-order terms occur then only those of even order may be nonzero. It is worth mentioning the assertion that in crystals, whose symmetry does not allow natural optical activity, the first terms in the expansion of the permittivity are quadratic terms [2]. Here we have proved that also the converse is true, namely, the occurrence of quadratic or higher-order terms rules out optical activity (first-order terms).

It is of interest to summarize the main results. For optically active media [Eqs. (4.1)], thermodynamics requires that [see Eqs. (4.2) and (4.9)]

$$\hat{\boldsymbol{\eta}}_2<\mathbf{0}, \quad \hat{\boldsymbol{\varepsilon}}_2>\mathbf{0}, \quad \hat{\boldsymbol{\alpha}}_2=-\hat{\boldsymbol{\xi}}_2^T, \quad \hat{\boldsymbol{\alpha}}_1=\hat{\boldsymbol{\xi}}_1^T, \quad \mathbf{N}=\mathbf{0}, \tag{6.1}$$

where $\hat{\boldsymbol{\xi}}=\hat{\boldsymbol{\mu}}^{-1}\hat{\boldsymbol{\lambda}}\hat{\boldsymbol{\varepsilon}}$ and $\hat{\boldsymbol{\eta}}=\hat{\boldsymbol{\mu}}^{-1}(\mathbf{1}-\omega^2\hat{\boldsymbol{\lambda}}\hat{\boldsymbol{\alpha}})$. Nondissipation means that $\hat{\boldsymbol{\varepsilon}}_2$ and $\hat{\boldsymbol{\eta}}_2$ vanish, namely,

$$\int_0^\infty \boldsymbol{\varepsilon}(u)\sin \omega u \; du=0, \quad \forall \omega\geqslant 0. \tag{6.2}$$

The condition (6.2) implies that the kernel $\boldsymbol{\varepsilon}(u)$ is zero for every $u\geqslant 0$. Only the constant value $\boldsymbol{\varepsilon}^0$ is allowed to be nonzero. In the isotropic case, α and λ may be complex valued, which means that the kernels of α and λ need not vanish. They vanish if the particular Drude-Born-Federov model is adopted.

For optically inactive media (higher-order terms), thermodynamics requires that [see Eqs. (5.3) and (5.4)]

$$\hat{\boldsymbol{\varepsilon}}_2>0, \; \hat{\boldsymbol{\mu}}_2>0, \; \hat{\boldsymbol{\gamma}}_2<0, \; \hat{\boldsymbol{\kappa}}_2<0, \tag{6.3}$$

while Eq. (5.2) holds. In terms of the functional representations, Eq. (6.3) means that the sine transform of the pertinent kernels are required to be definite, namely,

$$\int_0^\infty \boldsymbol{\varepsilon}(u)\sin \omega u \; du>0, \quad \int_0^\infty \boldsymbol{\gamma}(u)\sin \omega u \; du<0, \tag{6.4}$$

and analogously for the kernels $\boldsymbol{\mu}(u),\boldsymbol{\kappa}(u)$. By Eq. (5.2) the kernels $\boldsymbol{\alpha}(u),\boldsymbol{\beta}(u),\boldsymbol{\lambda}(u),\boldsymbol{\nu}(u)$ vanish for all $u\geqslant 0$.

The present results about optically active and optically inactive materials, with anisotropy and dissipation, are essentially different. They generalize the inequalities for the permittivity and the permeability of dispersive media that are derived by requiring that the divergence of the Poynting vector be negative (see [2], Chap. 2). The generality of the thermodynamic condition (2.7) is at the basis of the progress. In this regard it is crucial that the entropy flux **N**, related to nonlocality, is allowed to occur.

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