Analytical solution of Maxwell's equations in lossy and optically active crystals

Haijun Yuan
Liquid Crystal Institute, Kent State University, Kent, Ohio 44242

Weinan E
Mathematics Department, Princeton University, Princeton, New Jersey 08544

Peter Palffy-Muhoray
Liquid Crystal Institute, Kent State University, Kent, Ohio 44242
(Received 27 August 1999)

Analytical solution of Maxwell's equations is obtained for general linear optical materials: lossy and optically active crystals. Explicit expressions are obtained for the dispersion relation and the propagating eigenmodes. In general, four rather than two distinct modes are present. The results are useful in describing light propagation in optically complex media.

PACS number(s): 42.25.Bs

Light propagation in isotropic materials and simple crystals is well understood classically in terms of solutions of Maxwell's equations [1]. There exists today considerable interest in both experimental and theoretical work on optically active and other optically complex materials [2–9]. Analytic solutions of Maxwell's equations in such materials are not readily available.

Light propagation in lossy uniaxial crystals is discussed in detail in Ref. [10]. The problem is more formidable in lossy biaxial crystals, especially in the case where the real and imaginary parts of the dielectric tensor are not co-diagonal, and in materials where both natural and magnetically induced optical activities are present. Analytic solutions of Maxwell's equation in lossy biaxial crystals are first given in the work of G. Szivessy [11] (the original work is in German; we are not aware of translations in existence); similar results have been obtained subsequently [12]. Light propagation in a special case of lossless biuniaxial materials with natural and magnetically induced optical activity is considered in Ref. [5]; the general solution even in the lossless case is not known.

Solutions of Maxwell's equations describe the optical eigenmodes of the system, obtained by solving the eigenvalue problem for the fields. The eigenvalues are inversely proportional to the square of the wave vector; the solution of the secular equation gives the dispersion relation. Since for the propagating modes the electric displacement is normal to the wave vector, the solution manifold is the plane normal to it. The secular equation for lossless materials is therefore biquadratic. For lossy materials, as has been shown [11], the eigenvectors span the space normal to a complex wave vector. For optically active materials, the dielectric tensor is a function of the wave vector, leading to a nonstandard eigenvalue problem which has not been solved. Our contribution is to solve this problem for the case when the dielectric tensor depends linearly on the wave vector.

In this paper, we give explicit analytical expression for the solutions to Maxwell's equation in the most general homogeneous linear optical media: the lossy optically active biaxial crystal. Since our formalism allows a simple and compact description, we expect our results to be useful in a variety of applications, such as modelling light propagation in dye doped liquid crystals and the optics of magneto-optical storage materials.

The dielectric tensor. The nature of the solutions of Maxwell's equations is determined by the optical response implicit in the dielectric tensor which relates the electric field $\boldsymbol{E}$ and displacement $\boldsymbol{D}$ in the constitutive relation. In an infinite macroscopically homogeneous medium, linear response gives [13,14]

$$D_i = \varepsilon_{ik}(\omega,\boldsymbol{k})E_k, \tag{1}$$

where ω and $\boldsymbol{k}$ are the angular frequency and the wave vector, and

$$\varepsilon_{ik}(\omega,\boldsymbol{k}) = \delta_{ik} + \int_0^\infty \int f_{ik}(\tau,\boldsymbol{\rho}) e^{i(\omega\tau - \boldsymbol{k}\cdot\boldsymbol{\rho})} d^3\rho\, d\tau,$$

where $f_{ik}(\tau,\boldsymbol{\rho})$ is the susceptibility in real space, where τ and $\boldsymbol{\rho}$ are time and position. The spatially and temporally nonlocal response leads to the dependence of the permittivity on the wave vector $\boldsymbol{k}$ and frequency ω. We note that in general ε_{ik} is complex, but is neither symmetric, nor Hermitian. The generalized principle of symmetry of kinetic coefficients [15] leads to the symmetry condition

$$\varepsilon_{ik}(\omega,\boldsymbol{k},\boldsymbol{B}) = \varepsilon_{ki}(\omega,-\boldsymbol{k},-\boldsymbol{B}), \tag{2}$$

where $\boldsymbol{B}$ denotes an external static magnetic field. Disscussions of the symmetry of the dielectric tensor elsewhere [16–18] are in agreement with Eq. (2). The dielectric tensor may be written approximately as [13]

$$\varepsilon_{ik}(\omega,\boldsymbol{k}) = \varepsilon_{ik}^{(0)}(\omega) + i\gamma_{ikl}(\omega)k_l, \tag{3}$$

which may be regarded as a Taylor's series expansion of ε_{ik} to first order in $\boldsymbol{k}$. As a consequence of Eq. (2), $\varepsilon_{ik}^{(0)}(\omega)$ is a complex symmetric second rank tensor, while γ_{ikl} is a complex third rank tensor, both independent of $\boldsymbol{k}$; the product $\gamma_{ikl}k_l$ is a second rank antisymmetric tensor which can be written as

$$\gamma_{ikl}k_l = e_{ikl}g_l,$$

where e_{ikl} is the Levi-Civita symbol, and g_l denotes the gyration pseudovector describing natural optical activity. Here g_l depends on $\boldsymbol{k}$ and can be written as a product of a pseudotensor g_{lm} and $\boldsymbol{k}$

$$g_l = g_{lm}k_m.$$

The pseudotensor g_{lm} depends on the properties of the medium, it is in general complex and need not be symmetric.

Symmetry considerations lead to a similar contribution from a static external magnetic field B_m, and one obtains the general expression for the dielectric tensor

$$\varepsilon_{ik}(\omega,\boldsymbol{k},\boldsymbol{B}) = \varepsilon_{ik}^{(0)}(\omega) + ie_{ikl}g_{lm}^{(1)}k_m + ie_{ikl}g_{lm}^{(2)}B_m, \quad (4)$$

where $g_{lm}^{(1)}$ is the pseudotensor responsible for the natural optical activity and $g_{lm}^{(2)}$ is the tensor responsible for induced magnetic optical activity or Faraday effect.

Solution of Maxwell's equations. For convenience, we use the notation where $\boldsymbol{A}\overline{\overline{\varepsilon}}\boldsymbol{b} \equiv \boldsymbol{A}\cdot\overline{\overline{\varepsilon}}\cdot\boldsymbol{b}$ yielding a scalar.

We start with the relation of Eq. (1)

$$\boldsymbol{D}(k,\omega) = \varepsilon_0\overline{\overline{\varepsilon}}(\boldsymbol{k},\omega)\boldsymbol{E}(\boldsymbol{k},\omega), \quad (5)$$

where ε_0 is the permittivity of free space. Maxwell's equations give

$$(\overline{\overline{I}} - \hat{\boldsymbol{k}}\hat{\boldsymbol{k}})\overline{\overline{\varepsilon}}^{-1}\boldsymbol{D} = \lambda^2\boldsymbol{D}, \quad (6)$$

where $\lambda = (\omega/ck)$ and c is the speed of light. $\hat{\boldsymbol{k}}$ is a unit vector along $\boldsymbol{k}$; since $\boldsymbol{k}$ may be complex, we define $\hat{\boldsymbol{k}} = \boldsymbol{k}/k$ where $k = \sqrt{\boldsymbol{k}\cdot\boldsymbol{k}}$.

The secular equation is

$$\det[(\overline{\overline{I}} - \hat{\boldsymbol{k}}\hat{\boldsymbol{k}})\overline{\overline{\varepsilon}}^{-1} - \lambda^2\overline{\overline{I}}] = 0,$$

and using a representation with the complex orthogonal basis $\hat{\boldsymbol{m}}$, $\hat{\boldsymbol{n}}$, and $\hat{\boldsymbol{k}}$, this gives at once for the modes with $\lambda \neq 0$,

$$\lambda^4 - (\hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}^{-1}\hat{\boldsymbol{m}} + \hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}^{-1}\hat{\boldsymbol{n}})\lambda^2 + (\hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}^{-1}\hat{\boldsymbol{m}})(\hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}^{-1}\hat{\boldsymbol{n}}) - (\hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}^{-1}\hat{\boldsymbol{n}})(\hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}^{-1}\hat{\boldsymbol{m}}) = 0. \quad (7)$$

On making use of the Cayley Hamilton theorem [19], it follows [20] that

$$\det(\overline{\overline{\varepsilon}})\lambda^4 + (\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}^2\hat{\boldsymbol{k}} - tr(\overline{\overline{\varepsilon}})\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{k}})\lambda^2 + \hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{k}} = 0. \quad (8)$$

This expression for the secular equation is our first result.

Next we consider the dependence of the dielectric tensor $\overline{\overline{\varepsilon}}$ on λ. In the most general case, the dielectric tensor can be written, see Eq. (4), as

$$\overline{\overline{\varepsilon}}(\omega,\boldsymbol{k},\boldsymbol{B}) = \overline{\overline{\varepsilon}}_s(\omega) + \overline{\overline{G}}(\omega,\boldsymbol{k}) + \overline{\overline{G}}_B(\omega,\boldsymbol{B}), \quad (9)$$

where

$$\overline{\overline{G}} = -i\frac{\omega/c}{\lambda}(\overline{\overline{\gamma}}\hat{\boldsymbol{k}})\times = -i\frac{1}{\lambda}(\overline{\overline{\Gamma}}_k\hat{\boldsymbol{k}})\times = -i\frac{1}{\lambda}\boldsymbol{g}_k\times,$$

$$\overline{\overline{G}}_B = -i(B\overline{\overline{\gamma}}_B\hat{\boldsymbol{B}})\times = -i(\overline{\overline{\Gamma}}_B\hat{\boldsymbol{B}})\times = -i\boldsymbol{g}_B\times,$$

and substitution into Eq. (8) gives [20], after some algebra,

$$\xi_4\lambda^4 + \xi_3\lambda^3 + \xi_2\lambda^2 + \xi_1\lambda + \xi_0 = 0, \quad (10)$$

where

$$\xi_4 = \det(\overline{\overline{\varepsilon}}_s) - \boldsymbol{g}_B\overline{\overline{\varepsilon}}_s\boldsymbol{g}_B,$$

$$\xi_3 = 2\boldsymbol{g}_B\overline{\overline{\varepsilon}}_s\boldsymbol{g}_k,$$

$$\xi_2 = \hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}_s^2\hat{\boldsymbol{k}} - tr(\overline{\overline{\varepsilon}}_s)\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{k}} - \boldsymbol{g}_k\overline{\overline{\varepsilon}}_s\boldsymbol{g}_k + [\boldsymbol{g}_B^2 - (\hat{\boldsymbol{k}}\overline{\overline{\Gamma}}_B\hat{\boldsymbol{B}})^2],$$

$$\xi_1 = 2[\boldsymbol{g}_k\cdot\boldsymbol{g}_B - (\hat{\boldsymbol{k}}\cdot\boldsymbol{g}_k)(\hat{\boldsymbol{k}}\cdot\boldsymbol{g}_B)],$$

$$\xi_0 = \hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{k}} + (\overline{\overline{\Gamma}}_k\hat{\boldsymbol{k}})^2 - (\hat{\boldsymbol{k}}\overline{\overline{\Gamma}}_k\hat{\boldsymbol{k}})^2,$$

and the dependence of the dielectric tensor on the wave vector has been taken into account. Our approach avoids the introduction of spurious roots into the secular equation, and the results show that when the dependence of the dielectric tensor on the wave vector is linear, the secular equation is at most quartic, which may be solved analytically.

The eigenvectors $\boldsymbol{D}$ are simply obtained from Eq. (6):

$$\boldsymbol{D}\cdot\hat{\boldsymbol{k}} = 0,$$

$$\frac{\boldsymbol{D}\cdot\hat{\boldsymbol{n}}}{\boldsymbol{D}\cdot\hat{\boldsymbol{m}}} = \frac{\det(\overline{\overline{\varepsilon}})\lambda^2 - \hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{n}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{k}} + \hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{k}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{n}}}{\hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{k}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{n}} - \hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{n}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}\hat{\boldsymbol{k}}} \quad (11)$$

$$= \frac{[\det(\overline{\overline{\varepsilon}}_s) - \boldsymbol{g}\overline{\overline{\varepsilon}}_s\boldsymbol{g}]\lambda^2 - \hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{n}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{k}} + (\hat{\boldsymbol{n}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{k}})^2 + (\boldsymbol{g}\cdot\hat{\boldsymbol{n}})^2}{\hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{k}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{n}} - \hat{\boldsymbol{m}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{n}}\hat{\boldsymbol{k}}\overline{\overline{\varepsilon}}_s\hat{\boldsymbol{k}} + (\boldsymbol{g}\cdot\hat{n})(\boldsymbol{g}\cdot\hat{m}) - i\hat{k}\overline{\overline{\varepsilon}}_s\boldsymbol{g}}, \quad (12)$$

where $\boldsymbol{g} = \boldsymbol{g}_k/\lambda + \boldsymbol{g}_B$.

The expressions (10), (12) constitute the general solution of Maxwell's equations in homogeneous media, and are our main result. We note that the coefficients of the linear and cubic terms in the quartic secular equation, Eq. (10), are nonvanishing if both natural and magnetically induced optical activities are present. In general, therefore, the secular equation has four distinct roots; and consequently, in general, four, rather than two, distinct optical eigenmodes exist.

To examine these four modes, we consider the simple example of an isotropic medium where both natural optical activity and Faraday effect are present. Here we write the dielectric tensor as

$$\overline{\overline{\varepsilon}} = \varepsilon\overline{\overline{I}} - i\gamma\boldsymbol{k}\times - i\gamma_B\boldsymbol{B}\times, \quad (13)$$

where ε, and γ_B are scalars, and γ is a pseudoscalar. Letting $\Gamma_k = (\omega/c)\gamma$ and $\Gamma_B = B\gamma_B$, it follows that

$$\xi_4=\varepsilon(\varepsilon^2-\Gamma_B^2),$$

$$\xi_3=-2\varepsilon\Gamma_k\Gamma_B(\hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{B}}),$$

$$\xi_2=-2\varepsilon^2-\varepsilon\Gamma_k^2+\Gamma_B^2[1-(\hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{B}})^2],$$

$$\xi_1=0,$$

$$\xi_0=\varepsilon.$$

and assuming further that $\hat{\boldsymbol{k}}$ is along $\hat{\boldsymbol{B}}$, the secular equation becomes

$$(\varepsilon^2-\Gamma_B^2)\lambda^4-2\Gamma_k\Gamma_B\lambda^3-(2\varepsilon+\Gamma_k^2)\lambda^2+1=0 \qquad (14)$$

which can be factored into the two quadratics with four solutions which constitute the dispersion relations

$$\frac{k_{1,2}}{k_0}=\frac{-\Gamma_k\pm\sqrt{\Gamma_k^2+4(\varepsilon-\Gamma_B)}}{2}, \qquad (15)$$

$$\frac{k_{3,4}}{k_0}=\frac{\Gamma_k\pm\sqrt{\Gamma_k^2+4(\varepsilon+\Gamma_B)}}{2}, \qquad (16)$$

where $k_0=\omega/c$. Substituting the eigenvalues into Eq. (11) gives the polarizations of the eigenmodes

$$\frac{\boldsymbol{D}\cdot\hat{n}}{\boldsymbol{D}\cdot\hat{m}}=\frac{i\left(\frac{1}{\lambda}\Gamma_k+\Gamma_B\right)}{\varepsilon^2\lambda^2-\Gamma_k^2-2\lambda\Gamma_k\Gamma_B-\lambda^2\Gamma_B^2-\varepsilon}$$

$$=i \quad \text{for } \lambda_{1,2}$$

$$=-i \quad \text{for } \lambda_{3,4}.$$

This indicates that four circularly polarized eigenmodes exist in this medium with four different velocities.

For a slightly more complex example, consider an isotropic material with an additional symmetric term proportional to $\boldsymbol{k}\cdot\boldsymbol{B}$ in the dielectric tensor to give

$$\overline{\overline{\varepsilon}}=(\varepsilon+\gamma_{kB}(\boldsymbol{B}\cdot\boldsymbol{k}))\overline{\overline{I}}-i\gamma\boldsymbol{k}\times-i\gamma_B\boldsymbol{B}\times$$

$$=\left(\varepsilon+\frac{1}{\lambda}\Gamma_{kB}(\hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{B}})\right)\overline{\overline{I}}-i\left(\frac{1}{\lambda}\Gamma_k\hat{\boldsymbol{k}}\times+\Gamma_B\hat{\boldsymbol{B}}\times\right), \qquad (17)$$

where Γ_k,Γ_B are as before and $\Gamma_{kB}=\gamma_{kB}B(\omega/c)$. The dielectric tensor is still of the general form of Eq. (3).

Assuming $\hat{\boldsymbol{k}}\cdot\hat{\boldsymbol{B}}=1$ as before, the solution is again obtained by factoring into quadratics with four solutions which constitute the dispersion relations

$$\frac{k_{1,2}}{k_0}=\frac{-(\Gamma_k-\Gamma_{kB})\pm\sqrt{(\Gamma_k-\Gamma_{kB})^2+4(\varepsilon-\Gamma_B)}}{2}, \qquad (18)$$

$$\frac{k_{3,4}}{k_0}=\frac{(\Gamma_k+\Gamma_{kB})\pm\sqrt{(\Gamma_k+\Gamma_{kB})^2+4(\varepsilon+\Gamma_B)}}{2}. \qquad (19)$$

If Γ_{kB} is complex, the absorption for two modes with propagating along $\hat{\boldsymbol{B}}$ will differ from the absorption of the modes propagating opposite to $\hat{\boldsymbol{B}}$. The above behavior corresponds to experimentally observed ''magnetochiral anisotropy'' and ''magnetochiral dichroism'' [2,3]. Other illustrative examples will be discussed elsewhere [20].

In conclusion, we have obtained analytic solutions to Maxwell's equations in general linear media. The explicit expressions are useful for understanding optical behavior in complex optical media where both natural and magnetical optical activities may be present.

This work was supported in part by the NSF under ALCOM Grant No. 80-DMR20147 and by the AFOSR under MURI Grant No. F49620-17-1-0014.

[1] J.D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1967).
[2] G.L.J.A. Rikken and E. Raupach, Phys. Rev. E **58**, 5081 (1998).
[3] G.L.J.A. Rikken and E. Raupach, Nature (London) **390**, 493 (1997).
[4] M.J. Gunning and R.E. Raab, J. Opt. Soc. Am. B **14**, 1692 (1997).
[5] H. Taouk, J. Opt. Soc. Am. A **14**, 2006 (1997).
[6] T. Verbiest, M. Kauranen, and A. Persoons, J. Opt. Soc. Am. B **15**, 451 (1998).
[7] G. Barbero and A. Strigazzi, J. Appl. Phys. **58**, 2544 (1985).
[8] J.W. Goodby, Science **231**, 350 (1986).
[9] T. Wagersreiter and S. Mukamel, J. Chem. Phys. **105**, 7995 (1996).
[10] M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1975).
[11] G. Szivessy, *Handbuch Der Physik*, edited by H. Geiger and K. Scheel (Verlag Von Julius, Springer, Berlin, 1928).
[12] G.N. Ramachandran and Ramaseshan, *Handbuch Der Physik*, edited by S. Flugge (Springer-Verlag, Berlin, 1961).
[13] L.D. Landau, E.M. Lifshitz, and L.P. Pitaevskii, *Electrodynamics of Continuous Media*, 2nd ed. (Pergamon Press, Oxford, 1984).
[14] V.M. Agranovich and V.L. Ginzburg, *Crystal Optics with Spatial Dispersion, and Excitons* (Springer, New York, 1984).
[15] L.D. Landau and E.M. Lifshitz, *Statistical Physics*, 3rd ed. (Pergamon Press, Oxford, 1980), Pt. 1.
[16] P.N. Butcher and D. Cotter, *The Elements of Nonlinear Optics* (Cambridge University Press, Cambridge, 1990).
[17] P.M. Chaikin and T.C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, 1995).
[18] Yu. A. II'inskii and L.V. Keldysh, *Electromagnetic Response of Material Media* (Plenum Press, New York, 1994).
[19] Carl E. Person, *Handbook of Applied Mathematics, Selected Results and Methods*, 2nd ed. (Van Nostrand Reinhold, New York, 1983).
[20] H. Yuan and P. Palffy-Muhoray (unpublished).