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Non-linear optics in liquid-crystalline phases
A 4 × 4 matrix formalism

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The 4 × 4 matrix formalism of Berreman has been extended to include non-linear optical terms. A non-linear source has to be added to the original homogeneous differential equation, giving an inhomogeneous equation that can be solved by standard procedures. The final expression can be given in a compact form by applying the Hamilton–Cayley theorem. The formalism is illustrated in the treatment of three-wave mixing in cholesteric phases.

1. Introduction

Since lasers as high-intensity light sources have become commonly available, the field of non-linear optics has grown very rapidly, in both the theoretical and the experimental sectors [1, 2]. One of the most promising fields from the practical point of view, as well as being of technological interest, is that of wave mixing. Among the materials with large non-linear optical parameters, liquid crystals offer many advantages concerning the fulfillment of the phase-matching condition [3].

A description of non-linear optical phenomena has been obtained by considering only the electric field vector in Maxwell's equations and considering plane waves at normal incidence [1]. For light propagation in anisotropic media other methods have been developed, which also take account of the magnetic field vector in order to obtain a better description at interfaces and oblique incidences [4]. Among these methods, the 4 × 4 matrix formalism of Berreman [5] is the most widespread and has been extensively applied to light propagation in liquid crystals [6]. In the present study this method is extended to include non-linear sources. The final expression, regardless of the existence of non-linear optical terms, can be obtained in closed form by applying the Cayley–Hamilton theorem. The procedure is illustrated by applying the formalism to three-wave mixing in cholesterics.

2. The 4 × 4 matrix formalism

The description and introduction of the formalism have been given elsewhere [4, 5] and will not be repeated here. Following standard procedures [1], the curl equations contain a new term, $\mathbf{P}^{\mathrm{NLS}}$, which describes the non-linear effects. After introducing this term and eliminating the time dependence, we obtain

$$\begin{aligned} \operatorname{curl} \mathbf{H} &= -\frac{i\omega}{c}\mathbf{M} - \frac{4\pi i\omega}{c}\mathbf{P}^{\mathrm{NLS}}, \\ \operatorname{curl} \mathbf{E} &= -\frac{i\omega}{c}\mathbf{M}. \end{aligned} \tag{1}$$

Defining

$$\xi = \frac{\omega n_0}{c} \sin \phi_0, \tag{2}$$

where n_0 is the refractive index of the ambient medium and ϕ_0 is the angle of incidence, the equation (1) can be written in matrix form as

$$\frac{d\psi}{dz} = \frac{i\omega}{c}(\boldsymbol{\Delta}\psi + 4\pi\boldsymbol{\Omega}^{\text{NLS}}), \tag{3}$$

where ψ is the 4×1 generalized field vector [4, 5] and the matrix $\boldsymbol{\Delta}$ is defined in terms of the elements of $\mathbf{M}$. The non-linear part $\boldsymbol{\Omega}^{\text{NLS}}$ is defined by

$$\left.\begin{aligned}
\Omega_1^{\text{NLS}} &= P_3^{\text{NLS}}\left[M_{56}M_{63} - \left(M_{53} + \frac{c\xi}{\omega}\right)M_{66}\right]\Big/D,\\
\Omega_2^{\text{NLS}} &= P_1^{\text{NLS}} + P_3^{\text{NLS}}(M_{61}M_{63} - M_{13}M_{66})/D,\\
\Omega_3^{\text{NLS}} &= P_3^{\text{NLS}}(M_{43}M_{66} - M_{46}M_{63})/\text{D},\\
\Omega_4^{\text{NLS}} &= P_2^{\text{NLS}} + P_3^{\text{NLS}}\left[\left(M_{26} - \frac{c\xi}{\omega}\right)M_{63} - M_{23}M_{66}\right]\Big/D,
\end{aligned}\right\} \tag{4}$$

where

$$D = M_{33}M_{66} - M_{36}M_{63}. \tag{5}$$

In general both matrices $\boldsymbol{\Delta}$ and $\boldsymbol{\Omega}$ are arbitrary functions of z, and equation (3) has no analytical solution. In many cases, however, the matrix $\mathbf{M}$ is a constant, independent of z, or may be transformed into one of this type by appropiate redefinition of the variables. In such cases equation (3) is readily integrable and leads to analytical solutions. Note that equation (3) is inhomogeneous, and the solutions will involve those of the homogeneous case, i.e. those without the non-linear source. The field vector ψ oscillates with the final mixed wavelength, while the non-linear source depends on all of the wavelengths to be mixed. The boundary condition thus imposes $\psi\ (t < 0) = 0$, i.e. there is no wave-mixing before the pulse.

One particular problem of the 4×4 matrix formalism deals with the final solution of equation (3). Because of the exponential matrix, obtaining an analytical solution can be cumbersome since the exponential has to be developed in an infinite power series. This can be accomplished in a compact form by applying the Cayley–Hamilton theorem [7]. Thus powers higher than three can be expressed in terms of the lower powers because the matrix $\boldsymbol{\Delta}$ must satisfy its own characteristic polynomial for the eigenvalues (see also [8]). The solution of the homogeneous case of equation (3) for $\mathbf{M}$ independence of z can be written as

$$\psi(z) = (c_0\mathbf{I} + c_1\mathbf{L} + c_2\mathbf{L}^2 + c_3\mathbf{L}^3)\psi(0), \tag{6}$$

where $\mathbf{I}$ is the identity matrix, $\mathbf{L} = i\omega z\boldsymbol{\Delta}$ and the $\{c_i\}$ must be determined from the equation system relating the characteristic polynomial and the eigenvalues.

3. Three-wave mixing in cholesterics

As an illustration of the formalism presented in the previous section, the case of three-wave mixing in a cholesteric liquid crystal ($\omega = \omega_1 + \omega_2 + \omega_3$) is now presented. Three-wave mixing in cholesteric media is the lowest order for non-linear phenomena without an external field and has been studied many times [2, 9]. The first step in the calculation involves the definition of the matrix **M**; the cholesteric medium can be described as a twisted dielectric given by

$$\varepsilon_{ij} = \mathrm{A}_{ik}\mathrm{A}_{jl}\varepsilon^0_{kl}, \tag{7}$$

where

$$\{\varepsilon^0_{ij}\} = \begin{bmatrix} \varepsilon^0_{11} & 0 & 0 \\ 0 & \varepsilon^0_{22} & 0 \\ 0 & 0 & \varepsilon^0_{33} \end{bmatrix} \tag{8}$$

and A_{ij} is the rotation matrix

$$\{A_{ij}\} = \begin{bmatrix} \cos\theta & -\sin\theta & 0 \\ \sin\theta & \cos\theta & 0 \\ 0 & 0 & 1 \end{bmatrix}, \tag{9}$$

where $\theta = kz$ and $k = 2\pi/p$. k is the rotation per unit distance along the optical axis and p is the pitch of the helix. Using the identity

$$\mathbf{R}\ \mathrm{curl}\ \mathbf{E} = \mathrm{curl}\ (\mathbf{RE}) - k\mathbf{SRE}, \tag{10}$$

with

$$\mathbf{S} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{11}$$

the matrix **M** can be written as

$$\mathbf{M} = \begin{bmatrix} \boldsymbol{\varepsilon} & k\mathbf{S} \\ -k\mathbf{S} & \mathbf{I} \end{bmatrix}. \tag{12}$$

The field vectors are

$$\left.\begin{aligned} \mathbf{E}' &= \mathbf{R}(kz)\mathbf{E}, \\ \mathbf{H}' &= \mathbf{R}(kz)\mathbf{H} \end{aligned}\right\} \tag{13}$$

and the non-linear source term is [3]

$$\mathbf{P}^{(3)\prime} = -4\pi\left(\frac{\omega}{c}\right)^2 \chi^{(3)\prime}\mathbf{E}'(z, \omega_1)\mathbf{E}'(z, \omega_2)\mathbf{E}'(z, \omega_3), \tag{14}$$

where a prime refers to the twisted system. Note that all tensors are independent of z, and equation (3) has an analytical solution. Equation (3) has to be solved first in its homogeneous form for each of the frequencies involved; the solutions have the form

$$\psi(\omega_i, z) = \exp\left(-i\omega_i z\boldsymbol{\Delta}\right) \psi(\omega_i, 0). \tag{15}$$

The final solution for the generated wave is given by

$$\psi(\omega, z) = \exp\left(-i\omega z\boldsymbol{\Delta}\right)\int \boldsymbol{\Omega}^{\mathrm{NLS}}(\omega_1, \omega_2, \omega_3, z)\exp\left(i\omega z\boldsymbol{\Delta}\right) dz. \tag{16}$$

Note that the integral affects only the field parts and the tensors appear just as constant multiplicative factors. After some algebra, equation (16) gives the usual phase-matching condition as found in [3].

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