Optical Wave-Packet Propagation in Nonisotropic Media

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We show that the propagation equation for the slowly varying envelope of the electric field in a homogeneous dispersive nonisotropic medium contains terms that rotate the 3D wave packet of an optical pulse propagating as an extraordinary wave about an axis perpendicular to the propagation vector. It also possesses Fresnel diffraction coefficients that depend not only on the refractive index but also its derivatives with respect to direction. An analytic expression for the slowly varying envelope is obtained for an initial Gaussian wave packet by keeping terms up to second order in the wave equation for the slowly varying envelope.

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Propagation of light pulses in nonisotropic media, and the concepts of phase velocity, group velocity, group velocity dispersion, walk-off, etc., are well understood [1–6]. In this Letter we present the results of a derivation and analysis of the propagation equation for an optical wave packet (WP) in a homogeneous dispersive nonisotropic medium. We point out the existence of additional terms in the propagation equation originating due to finite beam size that can rotate the WP of an optical pulse propagating as an extraordinary wave about an axis perpendicular to the propagation vector. Furthermore, the propagation equation possesses Fresnel diffraction coefficients that depend not only on the refractive index but also its derivatives with respect to direction. For example, in a uniaxial crystal, cross terms of the form $\partial^2/\partial x \partial t$ are present in the wave equation for the slowly varying envelope (SVE), where x is the coordinate perpendicular to the direction of propagation $\mathbf{s}_0$ and in the plane formed by the optic axis and $\mathbf{s}_0$ (we take $\mathbf{s}_0$ as the $\mathbf{z}$ axis). This term rotates the WP about the y axis. Moreover, the Fresnel diffraction coefficients for the x and y coordinates are not equal; their difference is due to the contribution of terms proportional to the derivatives of the refractive index with respect to the angle ϑ between $\mathbf{s}_0$ and the optic axis. We derive an analytic expression for the SVE for an initial Gaussian pulse by keeping terms up to second order in the wave equation, and illustrate the dynamics of the propagation by presenting numerical examples in the uniaxial crystals beta-barium borate (BBO) and rutile (TiO_2).

The wave equation for the electric field $\mathbf{E}$ (in Gaussian units) in a homogeneous dispersive nonisotropic medium, $\nabla \times \nabla \times \mathbf{E} + (1/c^2)(\partial^2/\partial t^2)\mathbf{E} = -(4\pi/c^2)(\partial^2/\partial t^2)\mathbf{P}$, with linear polarization $\mathbf{P}$ given by $P_i(\omega, \mathbf{s}) = (4\pi)^{-1}[\varepsilon_{ij}(\omega, \mathbf{s}) - \delta_{ij}]E_j(\omega, \mathbf{s})$ ensures that the electric field can be written in as [1–5]

$$\mathbf{E}(\mathbf{x}, t) = (2\pi)^{-3} \times \int_{-\infty}^{\infty} d^3\mathbf{k}\, \mathbf{A}(\mathbf{k}) \exp\{i[n(\mathbf{k})\mathbf{k} \cdot \mathbf{x} - \omega(\mathbf{k})t]\}, \tag{1}$$

where $\omega(\mathbf{k}) = kc$ is the frequency of the wave component with (vacuum) wave vector $\mathbf{k}$, $\mathbf{s}$ is the unit vector in the $\mathbf{k}$ direction, $n(\omega, \mathbf{s})$ is the refractive index satisfying the secular equation $|\varepsilon_{ik} - n^2(\delta_{ik} - s_i s_k)| = 0$, ε is the dielectric tensor, and where we have taken the magnetic permeability equal to unity. Here $\mathbf{k} = (\omega/c)\mathbf{s}$ is not the wave vector $\mathbf{K}$ in the medium $[\mathbf{K} = n(\omega, \mathbf{s})(\omega/c)\mathbf{s}]$. Writing the electric field of a light pulse of central frequency ω_0 and central direction $\mathbf{s}_0$ in terms of the SVE $\mathbf{A}(\mathbf{x}, t)$ in the form [3,4],

$$\mathbf{E}(\mathbf{x}, t) = \mathbf{A}(\mathbf{x}, t) \exp\{i[n(\mathbf{k}_0)\mathbf{k}_0 \cdot \mathbf{x} - \omega(\mathbf{k}_0)t]\} + \text{c.c.}, \tag{2}$$

we find that the SVE can be written as

$$\begin{aligned} \mathbf{A}(\mathbf{x}, t) &= (2\pi)^{-3} \int_{-\infty}^{\infty} d^3\mathbf{k}\, \mathbf{A}(\mathbf{k}) \exp\left(i\{[n(\mathbf{k})\mathbf{k} - n(\mathbf{k}_0)\mathbf{k}_0] \cdot \mathbf{x} - [\omega(\mathbf{k}) - \omega(\mathbf{k}_0)t]\}\right), \\ &= (2\pi)^{-3} \int_{-\infty}^{\infty} d\omega \int d^2\mathbf{s}\, \mathbf{A}(\mathbf{k}) \exp\left(i\{[n(\omega, \mathbf{s})(\omega/c)\mathbf{s} - n(\omega_0, \mathbf{s}_0)(\omega_0/c)\mathbf{s}_0] \cdot \mathbf{x} - (\omega - \omega_0)t\}\right). \end{aligned} \tag{3}$$

The propagation equation in a nonisotropic dispersive medium for a finite size (non-plane-wave) pulse can be derived by considering the expression for the derivative of the SVE with respect to the spatial coordinate in the direction of $\mathbf{s}_0$. The factor $i[n(\omega, \mathbf{s})(\omega/c)\mathbf{s} - n(\omega_0, \mathbf{s}_0)(\omega_0/c)\mathbf{s}_0] \cdot \mathbf{s}_0$ in the expression for $\partial \mathbf{A}/\partial z$ can be expanded in a Taylor series in powers of $(\omega - \omega_0)$, $(\theta - \theta_0)$, and $(\phi - \phi_0)$. Then the replacement $(\omega - \omega_0)$ by $i(\partial/\partial t)$ to express terms involving powers of $(\omega - \omega_0)$ as time derivatives of corresponding order, and the systematic replacement of powers of $(\theta - \theta_0)$ and $(\phi - \phi_0)$ by the derivatives with respect to x and y can be made to obtain the propagation equation [7]. If we define the coordinate system so that the $\mathbf{k}_0$ is along z, the wave equation for the SVE in a general biaxial crystal is given by

$$\frac{\partial}{\partial z}\mathbf{A}(\mathbf{x},t) = \Big[-\beta_1\frac{\partial}{\partial t} - \frac{i}{2}\beta_2\frac{\partial^2}{\partial t^2} + \frac{1}{6}\beta_3\frac{\partial^3}{\partial t^3} + \cdots + \gamma_x\frac{\partial}{\partial x} + \gamma_y\frac{\partial}{\partial y} - \frac{i}{2}\gamma_{xx}\frac{\partial^2}{\partial x^2} - \frac{i}{2}\gamma_{yy}\frac{\partial^2}{\partial y^2} + i\gamma_{xt}\frac{\partial}{\partial x}\frac{\partial}{\partial t} + i\gamma_{yt}\frac{\partial}{\partial y}\frac{\partial}{\partial t} + i\gamma_{xy}\frac{\partial}{\partial x}\frac{\partial}{\partial y}\cdots\Big]\mathbf{A}(\mathbf{x},t)\,. \tag{4}$$

Details of the derivation of the wave equation and the dependence of the coefficients appearing in it on frequency and propagation direction will be presented elsewhere [7]. Here, the dispersion parameters $\beta_1 = [n(\omega_0,\mathbf{s}_0) + \omega_0\partial n(\omega_0,\mathbf{s}_0)/\partial\omega]/c$, $\beta_2 = d\beta_1/d\omega$, $\beta_3 = d\beta_2/d\omega$, are well known; the γ parameters will be described below. For a uniaxial crystal, the coefficients γ_y, γ_{yt}, and γ_{xy} vanish in our coordinate system.

The first term on the right-hand side of Eq. (4) specifies the projection of the group velocity along $\mathbf{s}_0$; β_1 is the inverse of the group velocity projection. The second term determines the group velocity dispersion (GVD); it increases the pulse duration and chirps the pulse as it propagates in the medium. The third term is a higher order dispersion term that can further increase the pulse duration, even if β_2 vanishes. The further terms vanish if the WP is a plane wave pulse of infinite extent propagating in the z direction. These additional terms change the propagation of the WP within the medium for a WP of finite extent. The fourth and fifth terms describe walk-off of the pulse; the coefficient γ_x (γ_y) is the ratio of the component of the group velocity along x (y) and $\mathbf{s}_0$. If we keep only the first order terms, Eq. (4) can be written in the form $(\mathbf{v}_g\cdot\nabla + \partial/\partial t)\mathbf{A}(\mathbf{x},t) = 0$ with $\mathbf{v}_g$ given by

$$\mathbf{v}_g = \frac{c}{[n(\omega_0,\mathbf{s}_0) + \omega_0\partial n(\omega_0,\mathbf{s}_0)/\partial\omega]} \times\Big[\mathbf{s}_0 - \frac{dn(\omega_0\theta_0)/d\theta}{n(\omega_0,\mathbf{s}_0)}\mathbf{e}_\theta - \frac{c}{n\sin\theta}dn(\omega,\theta,\phi)/d\phi\,\mathbf{e}_\phi\Big]. \tag{5}$$

The sixth and seventh terms describe diffraction of the WP in the directions perpendicular to $\mathbf{s}_0$. In a nonisotropic medium additional terms in the coefficients γ_{xx} and γ_{yy} proportional to $\partial n(\omega_0,\mathbf{s}_0)/\partial\theta$, $\partial n(\omega_0,\mathbf{s}_0)/\partial\phi$, $\partial^2 n(\omega_0,\mathbf{s}_0)/\partial\theta^2$, and $\partial^2 n(\omega_0,\mathbf{s}_0)/\partial\phi^2$ modify the Fresnel diffraction coefficient, $c/\omega_0 n(\omega_0)$, appropriate for isotropic dispersive media (see below). The γ_{xt} and γ_{yt} terms of Eq. (4) containing the mixed derivatives $\partial^2/\partial x\partial t$ and $\partial^2/\partial y\partial t$ are responsible for a number of effects. They rotate the WP about the y and x axes, respectively, increase the temporal width of the WP (in addition to the increase due to GVD), increase the dispersion in the x and y directions, respectively, and increase the chirp (in addition to that due to β_2) when β_2 and/or γ_{xx} and γ_{yy} are nonvanishing. The γ_{xy} term rotates the WP in the x-y plane. These effects are illustrated below.

If we keep terms only up to second order in Eq. (4), an analytic solution can be obtained for an initial Gaussian pulse. In a uniaxial crystal,

$$\mathbf{A}(\mathbf{x},t) = \mathbf{A}_0\frac{\exp\Big\{\frac{-[x + \gamma_x z - i\gamma_{xt}z\tau/(\tau_0^2 - i\beta_2 z)]^2}{2[\sigma_x^2 - i\gamma_{xx}z + \gamma_{xt}^2 z^2/(\tau_0^2 - i\beta_2 z)]}\Big\}\exp\Big[\frac{-y^2}{2(\sigma_y^2 - i\gamma_{yy}z)}\Big]\exp\Big[\frac{-\tau^2}{2(\tau_0^2 - i\beta_2 z)}\Big]}{\sqrt{[\sigma_x^2 - i\gamma_{xx}z + \gamma_{xt}^2 z^2/(\tau_0^2 - i\beta_2 z)](\sigma_y^2 - i\gamma_{yy}z)(\tau_0^2 - i\beta_2 z)}}, \tag{6}$$

where $\tau = t - \beta_1 z$, σ_x, σ_y, and $\sigma_z = \tau_0/\beta_1$ are the dispersions in x, y, and z, respectively, $\mathbf{A}_0$ lies in the optic plane, and we have taken the center of the initial WP at $(x,y,z) = (0,0,0)$. The propagation effects discussed in the previous paragraph can be understood from this expression. Substituting the form of the variable τ into Eq. (6), it is easy to see that the center of the WP moves with the group velocity which has a component β_1^{-1} in the z direction and a component $\gamma_x\beta_1^{-1}$ (the walk-off group velocity) in the x direction. The field temporally spreads and is chirped due to the presence of β_2 in the last term of the numerator of Eq. (6). The beam spreads in x and y due to the γ_{xx} and γ_{yy} terms in the first and second terms of the numerator, respectively. The

Fresnel diffraction coefficients γ_{xx} and γ_{yy} are modified by the anisotropy of the medium and are in general not equal [see Eqs. (8) and (9)]. The γ_{xt} term can rotate the WP about the y axis if $\gamma_{xx}z$ is not negligible compared to σ_x^2 or if $\beta_2 z$ is comparable to τ_0^2. This is evident from the sum of the exponents of the first and third terms in the numerator of Eq. (6) which is of quadratic form, $Ax^2 + B\tau^2 + Dx\tau$, where the coefficient D is proportional to γ_{xt}. The absolute value of the quadratic is a rotated ellipse in $x - c\tau$ space with rotation angle α, where $\tan 2\alpha = \mathrm{Re}(D/c)/\mathrm{Re}(A - B/c^2)$. The γ_{xt} term also increases the temporal width of the WP as it propagates in the medium. Moreover, it modifies the chirp across the pulse when β_2 is nonvanishing or when γ_{xx} is nonvanishing. Furthermore, the dispersion in the x direction increases quadratically with z due to the term containing $\gamma_{xt}^2 z^2$. For a uniaxial crystal the γ coefficients are given by the expressions

$$\gamma_x = -\frac{\partial n(\omega_0, \mathbf{s}_0)/\partial\theta}{n(\omega_0, \mathbf{s}_0)}, \tag{7}$$

$$\gamma_{xx} = \frac{c}{\omega_0 n(\omega_0, \mathbf{s}_0)}\left\{\frac{\partial^2 n(\omega_0, \mathbf{s}_0)/\partial\theta^2}{n(\omega_0, \mathbf{s}_0)} - 2\left[\frac{\partial n(\omega_0, \mathbf{s}_0)/\partial\theta}{n(\omega_0, \mathbf{s}_0)}\right]^2 - 1\right\}, \tag{8}$$

$$\gamma_{yy} = \frac{c}{\omega_0 n(\omega_0, \mathbf{s}_0)}\left\{\frac{\partial n(\omega_0, \mathbf{s}_0)/\partial\theta}{n(\omega_0, \mathbf{s}_0)} ctg(\theta_0) - 1\right\}, \tag{9}$$

$$\gamma_{xt} = -\frac{1}{n(\omega_0, \mathbf{s}_0)}\left\{\frac{\partial^2 n(\omega_0, \mathbf{s}_0)}{\partial\omega\,\partial\theta} - \frac{1}{n(\omega_0, \mathbf{s}_0)} \times \frac{\partial n(\omega_0, \mathbf{s}_0)}{\partial\omega}\,\frac{\partial n(\omega_0, \mathbf{s}_0)}{\partial\theta}\right\}. \tag{10}$$

The γ coefficients contain additional $\partial n(\omega_0, \mathbf{s}_0)/\partial\phi$ terms for the biaxial case.

In our first example we propagate an initial Gaussian WP with $\lambda_0 = 2\pi c/\omega_0 = 206$ nm, θ_0 (i.e., ϑ) $= 40°$, spatial widths $\sigma_x = 1.0\ \mu$m and $\sigma_y = \infty$, and temporal width $\tau_0 = 33.3$ fs propagating as an extraordinary ray in the negative uniaxial crystal BBO. After evaluating the coefficients appearing in Eq. (4), the propagation is computed using Eq. (6). Figure 1 shows the magnitude of the SVE $|\mathbf{A}(\mathbf{x}, t)|$ versus ct and x in a coordinate frame moving with the center of the pulse (the walk-off is in the negative x direction for BBO). Four plots are shown with $z = 0$ (the initial pulse), 1.0, 2.0, and 4 mm. The initial WP is 10 times broader in ct than in x. After propagating 1 mm, the pulse has significantly broadened in x due to Fresnel diffraction (γ_{xx}) and is roughly as broad in x as in the longitudinal distance ct; therefore the tilt of the WP is difficult to see in Fig. 1(b) (Fig. 2 shows that the contours of the WP are rotated ellipses, not circles). By $z = 2.0$ mm, the pulse has continued to broaden and the tilt of the WP is clearly evident; this trend continues through $z = 4.0$ mm. Figure 2 shows (a) the rotation angle α and (b) the major to semimajor axes ratio $\Re$ of the ellipse $\mathrm{Re}[Ax^2 + (B/c^2)(c\tau)^2 + (D/c)xc\tau]$ describing the contour lines of $|\mathbf{A}(\mathbf{x}, t)|$. The counterclockwise rotation of the ellipse increases with decreasing σ_x and with increasing z. The ellipse initially has $\alpha = 0$ [relative to the z axis—see Fig. 1(a)] but eventually becomes broader in x than z (i.e., $\alpha > 45°$) and the WP tilts; asymptotically for large z and $\sigma_x = 1.0\ \mu$m, $\alpha \approx 86.4°$ so the tilt of the WP relative to the x axis is 13.6°, whereas for $\sigma_x = 2.0\ \mu$m the tilt relative to the x axis is about 15.3°. The γ_{xt} coefficient responsible for the rotation is largest for frequencies closest to the resonance frequencies of the medium and for angles ϑ far from 0° or 90° (this determined our choice of frequency and ϑ). The ratio $\Re$ initially dramatically decreases as the width in x increases due to Fresnel diffraction, and eventually exceeds the width in $z(ct)$. $\Re$ goes through a minimum, and the value and location of the minimum depend on σ_x. The $z \to \infty$ limits of α and $\Re$ can be determined analytically but the expressions are complicated; we do not present them here.

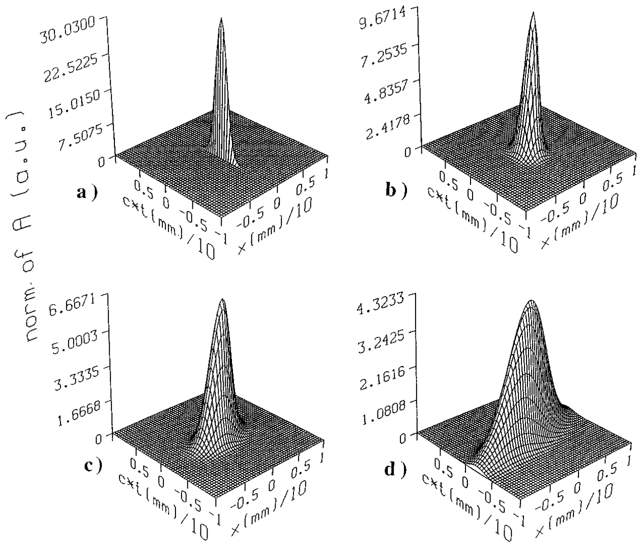

FIG. 1. $|A(\mathbf{x}, t)|$ for an initial extraordinary Gaussian wave packet with $\theta_0 = 40°$, $\lambda_0 = 2\pi c/\omega_0 = 206$ nm, spatial width $\sigma_x = 10\ \mu$m, temporal width $\tau_0 = 33.3$ fs, and $\sigma_y = \infty$ in BBO versus ct and x in the frame moving with the center of the pulse for (a) $z = 0$, (b) $z = 0.1$ mm, (c) 2.0, and (d) 4 mm.

As a second example we consider propagation of an initial Gaussian WP with $\lambda_0 = 2\pi c/\omega_0 = 410$ nm, $\theta_0 = 40°$, spatial widths $\sigma_x = 1.0\ \mu$m and $\sigma_y = \infty$, and temporal width $\tau_0 = 75$ fs propagating as an extraordinary ray in the positive uniaxial rutile crystal. Figure 3 shows contour plots of $|A(\mathbf{x}, t)|$ versus ct and x for $z = 0$, 1, 2, and 4 mm. Now, the direction of rotation of the ellipse is clockwise (and the walk-off is in the

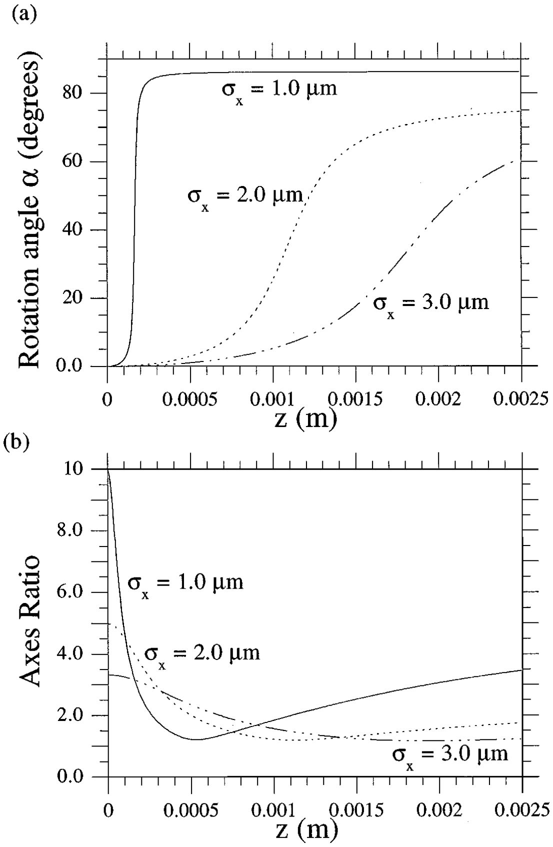

FIG. 2. (a) Rotation angle α and (b) major to semimajor axes ratio $\Re$ of the ellipse describing the contour lines of $|A(\mathbf{x}, t)|$ as a function of z for three different values of the initial spatial width σ_x. The value of the other parameters are as in Fig. 1.

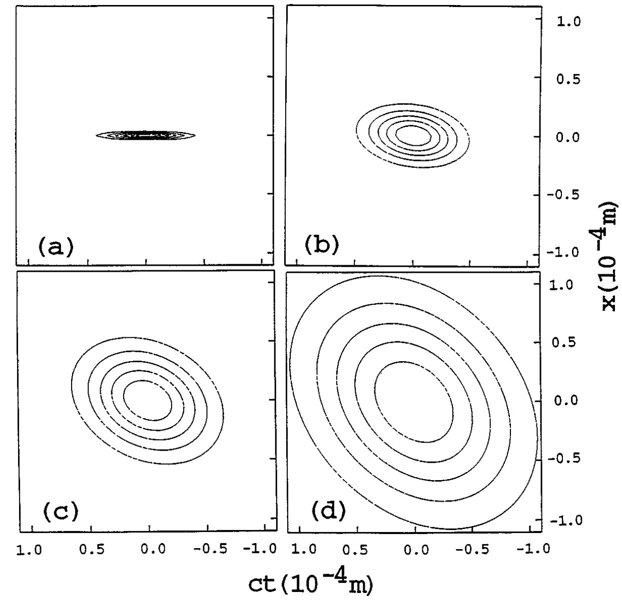

FIG. 3. Contour plots of $|A(\mathbf{x}, t)|$ for an initial extraordinary Gaussian wave packet with $\theta_0 = 50°$, $\lambda_0 = 2\pi c/\omega_0 = 410$ nm, spatial width $\sigma_x = 1.0$ μm, temporal width $\tau_0 = 75$ fs, and $\sigma_y = \infty$ in rutile versus ct and x in the frame moving with the center of the pulse for (a) $z = 0$, (b) $z = 0.1$ mm, (c) 2.0, and (d) 4 mm.

positive x direction). Otherwise, the same general trends for α and $\Re$ occur for rutile as for BBO.

In summary, we developed a method for determining the propagation equation for the SVE of the electric field in a homogeneous dispersive nonisotropic medium and for computing the coefficients appearing in it. The propagation equation contains terms that rotate the WP of an optical pulse about an axis perpendicular to the propagation vector, and it possesses Fresnel diffraction coefficients that depend not only on the index of refraction but also its derivatives with respect to direction. We used the analytic solution for the SVE for an initial Gaussian WP to demonstrate these points in BBO and rutile crystals. This work has application in such diverse fields as nonlinear optics, near field optical microscopy in nonisotropic media, and light pulse propagation in the intergalactic medium which is nonisotropic due to magnetic fields.

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