Nonlinear optics

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March 3, 2014

Abstract

In the laser physics course, nonlinear effects have only briefly been treated. Interesting effects arise when the polarization of a medium depends nonlinearly on the electric field. Two such effects, the Kerr effect and three wave mixing, have been investigated here. The Kerr effect can be used to make self-focusing lenses. Three wave mixing can be used to add, subtract or double the frequency of laser beams.

In this work, it was shown how these effects arise from the third and second order terms in the Taylor expansion of the polarization.

A numerical simulation of beam self-focusing using the Kerr effect was attempted. Due to stability problems, however, no results of this were obtained.

Contents

1 Introduction

During the better part of the course we have studied linear phenomena, that is the interaction between matter and light described by the wave equation

$$\nabla^2 \mathbf{E} - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 \mathbf{P}}{\partial t^2}, \tag{1}$$

where the polarization $\mathbf{P}$ is linear wrt. the electric field $\mathbf{E}$, as described by $\mathbf{P} = \epsilon_0 \chi \mathbf{E}$, where χ is electric susceptibility of the medium and ϵ_0 is the vacuum permittivity. The course briefly touched upon nonlinear phenomena, where the polarization is taylor expanded as $\mathbf{P} = \epsilon_0(\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{E}^2 + \dots)$.

In this project, we describe and discuss two effects of such a nonlinearity: beam self-focusing using the Kerr effect; and wave mixing.

2 The Kerr effect

The Kerr effect is found in media where the index of refraction depends on the electric field. For practical laser beams, the amplitude of the electric field depends on the position in the beam. This leads to spatial variation of the index of refraction. Under certain and experimentally obtainable conditions, this gives rise to beam self-focusing within a passive medium.

2.1 Mathematical details at a glance

The mathematical formulation of the Kerr effect is derived in **milonni**. For a monochromatic laser field $E = \mathcal{E}(x, y, z)e^{-i\omega t}$ with frequency ω in an isotropic and centrosymmetric medium (i.e. a medium which quantum states have definite parity), an alternative wave equation for the complex laser field and polarization amplitudes is found. The most important point here is, that the polarization has a linear and a nonlinear contribution $\mathcal{P} = \mathcal{P}^{\mathrm{L}} + \mathcal{P}^{\mathrm{NL}}$:

$$\nabla^2 \mathcal{E} + \frac{\omega^2}{c^2}\mathcal{E} = -\frac{\omega^2}{\epsilon_0 c^2}\mathcal{P} = -\frac{\omega^2}{\epsilon_0 c^2}\left(\epsilon_0 \chi_1 \mathcal{E} + \epsilon_0 \chi_3 |\mathcal{E}|^2 \mathcal{E}\right). \tag{2}$$

It should be noted, that the susceptibilities here are not simple scalars, but are certain values of the nonlinear susceptibility tensor. For a monochromatic field, χ_1 and χ_3 are constant as ω is constant. Defining

$$-\chi \frac{\omega^2}{c^2}\mathcal{E} \equiv -\chi_1 \frac{\omega^2}{c^2}\mathcal{E} - \chi_3 \frac{\omega^2}{c^2}|\mathcal{E}|^2\mathcal{E} \tag{3}$$

FiXme Warning: Implicit approximation: feltet er ikke så stærkt, at der kræves højere ordener.

and using $n^2 = 1 + \chi$ yields

$$n = \sqrt{1 + \chi_1 + \chi_3|\mathcal{E}|^2} = n_0\sqrt{1 + \frac{\chi_3}{n_0^2}|\mathcal{E}|^2} \tag{4}$$

$$\simeq n_0 + \frac{\chi_3}{2n_0}|\mathcal{E}|^2 \equiv n_0 + \tfrac{1}{2}n_2|\mathcal{E}|^2, \tag{5}$$

where $n_0 = \sqrt{1 + \chi_1}$ is the linear refractive index in the medium. It is seen that the effective refractive index depends quadratically on the electric field amplitude $\mathcal{E}(x, y, z)$.

2.2 Qualitative interpretation and uses

As shown in figure 1, a Gaussian beam, which is a beam exhibiting cylindrical symmetry and where the intensity distribution is or nearly is Gaussian, feels a larger refractive index closer to the beam axis. Eqn. (5) shows that this nonlinear correction is proportional to the norm squared of the field amplitude, i.e. the beam intensity.

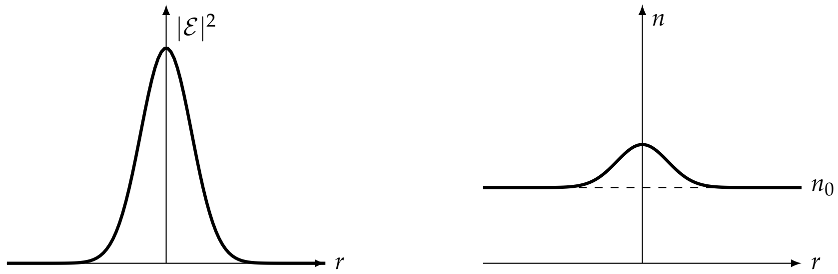


Figure 1 Gaussian beam profile (left) and effective refractive index (right) in arbitraty units. Note, that the contribution to the refractive index due to the Kerr effect $\frac{1}{2}n_2|\mathcal{E}|^2$, is not to scale wrt. the linear refractive index n_0.

The beam size of a Gaussian beam is not constant, but varies along the beam axis z, with minimum beam cross section found at the beam waist. Using a geometric interpretation of the light, rays moving away from the waist have increasing distance from the axis, i.e. radial distance r, leading to a broadening of the beam and a lower intensity. As such, the rays move from a region of high to lower refraction index, leading to a reflection of the ray back towards the beam axis.

This self-focusing effect competes with diffractive spreading. Below a certain critical intensity, self-focusing is not observed. This limit is normally given in terms of total beam power, for example:

$$P_{\text{crit}}^{\text{gauss}} \sim \frac{c\epsilon_0}{8\pi}\frac{\lambda^2}{n_2} \qquad \text{Gaussian beam [\textbf{milonni}],} \tag{6}$$

$$P_{\text{crit}}^{\text{cyl}} \approx \frac{ca^2}{64}\frac{\lambda^2}{n_2} \qquad \text{Cylindrical beam of uniform intensity [\textbf{prl-selftrap}],} \tag{7}$$

where the angular diffractive divergence of the cylindrical beam is $a\lambda/n_0 2r$. Note here, that the critical power decreases linearly in n_2 but increases quadratically in wavelength.

As can be expected, extreme power past the critical power will lead to undesired effects in the medium. These effects include multi photon ionisation and other dielectric breakdown effects, potentially leading to destruction of the medium.

In optics, media exhibiting this self-focusing effect are called Kerr lenses, as the passive medium acts as a lens for beams passing through. The dioptric power (inverse focal length) of a Kerr lens of length z for a circular Gaussian beam of power P with waist radius w_0 is [**yefet-kerrlens**]

$$f_{\text{kerr}}^{-1} = \frac{4}{\pi}\frac{n_2 P}{w_0^4}z. \tag{8}$$

Kerr lenses have the added advantage over standard optical lenses, that the Kerr lenses do not require alignment of the optical axis of the lens and the beam axis. This is due to the fact that the optical axis of the Kerr lens is induced by the electric field of the laser beam. This advantage is utilized in Kerr lens mode-locking.

2.3 Numerical work

We wish to confirm the numerical results from **prl-selffocus**. That is, we wish to show that a Gaussian beam is focused when propagating in a Kerr lens. Our starting point is the full wave equation, and the assumption that the propagating beam is cylindrical. Per **prl-selffocus** this leads to a dimensionless form of the wave equation

$$i\frac{\partial \tilde{E}}{\partial \tilde{z}} + \frac{\partial^2 \tilde{E}}{\partial \tilde{r}^2} + \frac{1}{\tilde{r}}\frac{\partial \tilde{E}}{\partial \tilde{r}} + |\tilde{E}|^2\tilde{E} = 0, \tag{9}$$

where $\tilde{r} = r/a$, a is a characteristic length of the beam intensity profile, $\tilde{z} = z/2ka^2$ is the characteristic axial length of the beam, $k = n_0\omega/c$ is the wave number of the laser field and $\tilde{E} = (\chi_1/\chi_3)^{1/2}ka\mathcal{E}$.

The point of solving the dimensionless wave equation is, that we minimize the dependence on physical properties of the beam. This simplifies the numerical approach and makes it easier to obtain a solution exhibiting the correct physical behaviour albeit for at generalized monochromatic circular Gaussian beam.

The partial differential equation given in Eqn. (9) is categorized as a 1D diffusion problem. To simplify notation, we drop the tildes from the variables in the following discussion, but the variables remain dimensionless. A method for solving this problem is the Crank-Nicolson method, which applies for equations of the form

$$\frac{\partial u}{\partial t} = F\Big(u, x, t, \frac{\partial u}{\partial x}, \frac{\partial^2 u}{\partial x^2}\Big). \tag{10}$$

This is the case for our equation, since it can be rewritten as

$$i\frac{\partial E}{\partial z} = -\frac{1}{r}\frac{\partial E}{\partial r} - \frac{\partial^2 E}{\partial r^2} - |E|^2 E. \tag{11}$$

In order to evaluate this numerically, we discretise z and r in a grid. The first and second order derivatives can then be approximated by the central finite difference

$$f'(x) = \frac{f(x+\frac{1}{2}h) - f(x-\frac{1}{2}h)}{h} \quad f''(x) = \frac{f(x+h) - 2f(x) + f(x-h)}{h^2}. \tag{12}$$

The Crank-Nicolson method relates the value of E at the $n+1$ z-step to the value in the n'th step via the equalities of the forward and backward Euler method

$$\frac{E_i^{n+1} - E_i^n}{\Delta z} = \frac{1}{2}\big[F_i^{n+1} + F_i^n\big]. \tag{13}$$

Note that i denotes the step in the r direction. The right hand side can be evaluated using the central difference approximation. This gives a set of equations for E^{n+1} that depends on E^n i.e. the values in the previous step

$$\begin{aligned}(\alpha + 2\beta^2 - |E_i^{n+1}|^2)E_i^{n+1} - \beta^2(E_{i+2}^{n+1} + E_{i-2}^{n+1}) - \frac{\beta}{r_i}(E_{i+1}^{n+1} - E_{i-1}^{n+1})\\ = (\alpha - 2\beta^2 + |E_i^n|^2)E_i^n + \beta^2(E_{i+2}^n + E_{i-2}^n) + \frac{\beta}{r_i}(E_{i+1}^n - E_{i-1}^n),\end{aligned} \tag{14}$$

where $\alpha = -2i/\Delta z$ and $\beta = 1/2\Delta r$.

This is almost a five-diagonal matrix equation except for the term that is cubic in E_i^{n+1}. However with sufficiently small steps one can approximate the cubic term so that $|E_i^{n+1}|^2 E_i^{n+1} \approx |E_i^n|^2 E_i^{n+1}$ i.e. take the quadratic part from the last step. In each time step, one must then solve the matrix inversion problem

$$\mathbf{A}\mathbf{E}^{n+1} = \mathbf{B}\mathbf{E}^n, \tag{15}$$

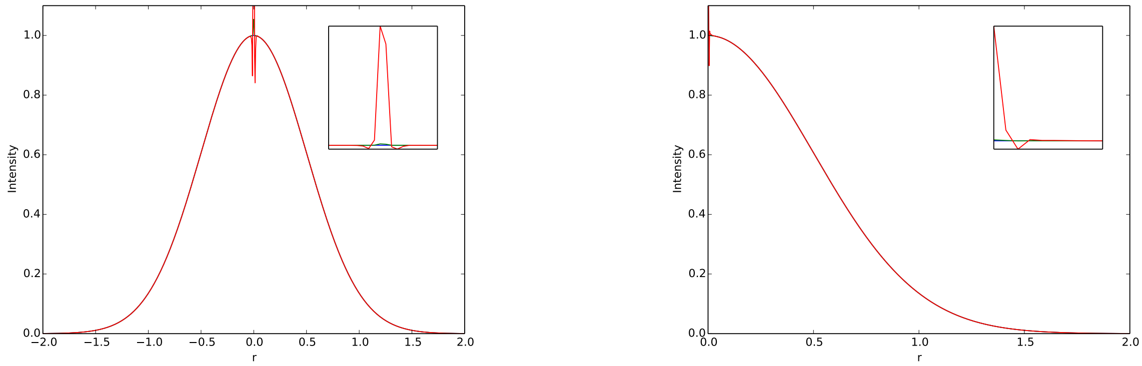
where the coefficients that make up these two matrices are the coefficients listed in Eqn. (14).

Results

We have implemented the Crank-Nicolson algorithm in the Python language. The source code can be found at https://github.com/Munken/Laser.

For the numerical simulation, the boundary conditions are quite important. Since our beam is Gaussian, we enforce that $E \to 0$ as $r \to \infty$. Furthermore the first derivative in $r = 0$ should be 0 since the beam is cylindrically symmetric.

We have tried to introduce these boundary conditions in two ways. Figure 2(a) shows a simulation where we have simulated the interval $[-r_{\max}, r_{\max}]$. Figure 2(b) shows the same simulation where we have only simulated from $r = 0$. It is clear from these two figures that we have numerical instabilities for



(a) Solution over the interval $[-r_{\max}, r_{\max}]$

(b) Solution over the interval $[0, r_{\max}]$

Figure 2 The result of our numerical simulation after propagating 10 steps. The insets shows the behaviour around $r = 0$. No sign of any change of the shape of the beam profile was observed.

the small r-values. We have not been able to solve this problem so we have not been able to reproduce **[prl-selffocus**][Fig. 1].

3 Three-wave mixing

Three-wave mixing is an effect arising from the second order nonlinearity of the polarity $\mathbf{P} = \epsilon_0(\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{E}^2 + \dots)$. In a medium with this type of nonlinearity, two incoming waves can couple together and form one or more additional waves. As the two waves induce an oscillating polarization in the material, the nonlinearity couples the frequencies together to the sum and difference frequencies. By the otherwise linear wave equation (1), these new polarization modes will couple back to the electric field, resulting in sum and difference frequency electromagnetic waves.

3.1 Mathematical derivation

To examine the consequences of the second order nonlinearity, let two plane waves of frequencies ω_1 and ω_2 with electric field amplitude $\mathcal{E}_1$ and $\mathcal{E}_2$, respectively, enter the nonlinear medium in superposition:

$$\mathbf{E}(t) = \Re\left(\mathcal{E}_1 e^{i(\mathbf{k_1}\cdot\mathbf{r}-\omega_1 t)} + \mathcal{E}_2 e^{i(\mathbf{k_2}\cdot\mathbf{r}-\omega_2 t)}\right).$$

The second order part of the polarity becomes[**saleh**]

$$\begin{aligned}\mathbf{P}^{NL} &= \varepsilon_0\chi^{(2)}\mathbf{E}^2(t)\\ &= \varepsilon_0\chi^{(2)}2\Re\left[\left(|\mathcal{E}_1|^2 + |\mathcal{E}_2|^2\right)e^0 + |\mathcal{E}_1|^2 e^{i2(\mathbf{k_1}z-\omega_1 t)} + |\mathcal{E}_2|^2 e^{i2(\mathbf{k_2}z-\omega_2 t)}\right.\\ &\left.+2\mathcal{E}_1\mathcal{E}_2 e^{i((\mathbf{k_1}+\mathbf{k_2})\cdot\mathbf{r}-(\omega_1+\omega_2)t)} + 2\mathcal{E}_1\mathcal{E}_2^* e^{i((\mathbf{k_1}-\mathbf{k_2})\cdot\mathbf{r}-(\omega_1-\omega_2)t)}\right].\end{aligned} \quad (16)$$

It is seen that new frequencies are generated: 0, the uninteresting[1] one, $2\omega_1$ and $2\omega_2$, which are second harmonics of the original frequencies, and more interestingly, the sum and difference of the initial frequencies. The last two frequency couplings are often referred to as up- and down-conversion respectively.

[1]Being constant, it drops out of the wave equation and produces no radiation.

It is clear from eq. (16) that for the up- and down-conversion, the third wave frequency and -vector will be:

$$\omega_3 = \omega_1 \pm \omega_2 \tag{17}$$
$$\mathbf{k}_3 = \mathbf{k}_1 \pm \mathbf{k}_2. \tag{18}$$

Once the third wave is created, it will also couple back to the two other waves, but these constraints secures that a neutral interaction is sustained through time and space. This effect will add to the spectrum, however.

3.2 Numerical formulation of up-conversion

To get a numerical interpretation of the result, one can assume the eq. (17) and inject the up-conversion part of eq. (16) into the time dependent wave equation of each wave [**shen**]:

$$\left(\frac{\partial}{\partial z} + \frac{1}{v_i^g}\right)\mathcal{E}_i\left(z,t\right) = \frac{i2\pi\omega_i^2}{kc^2}\mathbf{P}^{NL}\left(z,t\right)e^{i(k_i z - \omega_i t)} \Rightarrow$$
$$\left(\frac{\partial}{\partial z} + \frac{1}{v_i^g}\right)\mathcal{E}_i\left(z,t\right) = \frac{i\omega_i\chi^{(2)}}{2n_i c}\mathcal{E}_j^*\mathcal{E}_k e^{i(k_3-k_2-k_1)z}, \text{where } i \neq j \neq k$$

where the identity $\omega_i = k_i c$ has been used, and also the rotating wave approximation and slowly varying amplitude approximation. These are equivalent to the coupled partial differential equations (2) in [**bakker**] and could be numerically solved with the Runge-Kutta procedure as described therein.

Typically this is done in KDP or KTP crystals such as the sum frequency conversion done in [**sumFreq**] where a 455 nm is made from a 807 nm and a 1062 source. Though only with an efficienty of 0.01%, which is assumed to be due to bad angle tuning. Efficiency's up to 93.7% has been achieved by DTU Fotonik [**DTU**], which they use for optical coherence tomography, because it provides the freedom to use a tunable laser as one of the mixing frequencies, such that a chirpable output can be achieved.

Multiple wave mixing can be optained at higher orders of nonlinearity terms, such that the third order gives rise to four-wave mixing and so on.

4 Conclusion

In this report the Kerr effect and wave mixing have been investigated.

The Kerr effect on the refractive index was worked out from the second nonlinear term of the polarization. From this the self-focusing effect could be explained by the in-homogeneous refractive index composition. And the critical intensity for this effect to conquer diffractive spreading was presented for the cylindrical and the Gaussian beam, which was basis wave for the numerical work.

Here the point of departure was the dimensionless waveequation, where the second order derivatives was approximated by the central finite difference. Then using the Crank-Nicolson method the problem was reformulated to a five diagonal matrix which should be solved for each time step. The implementation of the Crank-Nicolson was numerically unstable. Though a tendency of centred amplification was observed, either energy conservation broke down, or strong oscillations manifested at the centre.

Because of the issues with the numerical solutions to the partial differential equations a qualitative explanation of three wave mixing was presented. The second order term couples the two waves together, which then couple back to the electric field creating a third wave. The frequency will be the sum or difference of the initial frequencies. Finally examples of the up-conversion in praxis with efficiency's up to 93.7% are presented.