University of Hamburg, Department of Physics Nonlinear Optics Kärtner/Mücke, WiSe 2019/2020 Problem Set 4

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1. An arbitrary wave polarized in the x-y-plane is described by

$$\vec{E}(z,t) = \frac{1}{2} \left[E_x(\omega) e^{j(\omega t - kz)} + c.c. \right] \hat{\mathbf{x}} + \frac{1}{2} \left[E_y(\omega) e^{j(\omega t - kz)} + c.c. \right] \hat{\mathbf{y}}$$

and propagates through an instantaneously reacting isotropic and lossless medium that shows a third-order nonlinearity.

Consider the generated nonlinear polarization at the third harmonic $P_x^{(3)}(3\omega)$.

(a) Show that the nonlinear polarization at the third harmonic can be expressed as:

$$P_x^{(3)}(3\omega) = \frac{\varepsilon_0}{4} \chi_{xxxx} E_x^3(\omega) + \frac{\varepsilon_0}{4} \left[\chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx} \right] E_y^2(\omega) E_x(\omega)$$

To solve the problem remember the mirror image symmetry of an isotropic medium mentioned in the course manuscript: in the third order susceptibility tensor an index cannot appear an odd number of times (each index must occur at least twice). You can start with the equation 2.4 of the manuscript.

(b) Show that in the isotropic medium the following relation is valid:

$$\chi_{xxxx} = \chi_{xxyy} + \chi_{xyxy} + \chi_{xyyx}$$

and show that the nonlinear polarization at the third harmonic can be expressed as:

$$P_x^{(3)}(3\omega) = \frac{\varepsilon_0}{4} \chi_{xxxx} \left[E_x^3(\omega) + E_y^2(\omega) E_x(\omega) \right]$$

To solve this part consider a general rotation tensor $R(\theta)$ around the z-axis:

$$R(\theta) = \begin{pmatrix} \cos(\theta) & \sin(\theta) & 0\\ -\sin(\theta) & \cos(\theta) & 0\\ 0 & 0 & 1 \end{pmatrix}$$

Using that matrix the rotation of the susceptibility tensor elements follows the same way as we see in the chapter 2.4.5 of the manuscript. Also remember the valid elements of the susceptibility tensor of an isotropic medium mentioned in the part a).

- (c) Show that circularly polarized light $(E_y = \pm jE_x)$ can not generate thirdharmonic light in this medium.
- (d) Now let's consider a third order process of the form $\omega = \omega_{in} + \omega_{in} \omega_{in}$. This basically means a polarization $P^{(3)}(\omega; \omega, \omega, -\omega)$ at frequency ω is induced via the third-order mixing of fields at the same frequency. Such process induces a change in refractive index for the driving field. Show that in an instantaneous, isotropic and lossless medium, $P_x^{(3)}(\omega)$ is given by the following expression:

$$P_x^{(3)}(\omega) = \frac{1}{4} \varepsilon_0 \chi_{xxxx} \left[3 |E_x|^2 E_x + 2 |E_y|^2 E_x + E_y^2 E_x^* \right]$$

Remember $E_i(-\omega) = E_i(\omega)^*$.

- (e) Derive the analogous expression for $P_y^{(3)}(\omega)$.
- (f) Derive an expression for $P_{\pm}^{(3)}(\omega)$ for the case of circularly polarized light. Circularly polarized light is described by:

$$E_{\pm} = \frac{1}{\sqrt{2}} \left(E_x \pm j E_y \right)$$

Give expressions for $P_{\pm}^{(3)}(\omega)$.

Hint: The polarization can be written in the same way as the electric field in circular components:

$$P_{\pm}^{(3)}(\omega) = \frac{1}{\sqrt{2}} \left(P_x^{(3)}(\omega) \pm j P_y^{(3)}(\omega) \right)$$
(1)

(g) The intensity-dependent refractive index n_2 is defined by:

$$n = n_0 + n_2 I$$

Give an expression for n_2 for the case of linearly and circularly polarized light.

To solve this problem remember the basic relations between the electric field, susceptibility and refractive index from chapter 1 of the manuscript:

$$P = \varepsilon_0 \chi E = \varepsilon_0 (n^2 - 1)E \tag{2}$$

Have a look into that chapter and see how a change Δn of the material refractive index can be dependent of the intensity of the wave.

(h) Give a physical interpretation of the expressions derived in problems e-g. Remember again, we have been dealing with the third order polarization during the whole problem.