Questions related to the first course
Introduction to nonlinear optics

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We consider the propagation of a monochromatic optical beam through a material. The nonlinear susceptibilities that characterize the medium are described by scalar quantities and their dispersions in frequencies are neglected. The amplitude of the electric field associated with the beam is described by a real quantity:

$$\boldsymbol{\mathcal{E}}(z,t) = \boldsymbol{E}(\omega)e^{-\imath\omega t} + \boldsymbol{E}(-\omega)e^{+\imath\omega t}.$$

Similarly, the macroscopic polarization emitted into the material at the frequency ω is written:

$$\boldsymbol{\mathcal{P}}(z,t) = \boldsymbol{P}(\omega)e^{-\imath\omega t} + \boldsymbol{P}(-\omega)e^{+\imath\omega t}.$$

1 Notations

1. In a general way, is the amplitude E described by a real quantity?
2. What is the relation between $\boldsymbol{E}(\omega)$ and $\boldsymbol{E}(-\omega)$?
3. Recall the relationship between $\boldsymbol{P}(\omega)$ and $\boldsymbol{E}(\omega)$ in a linear and isotropic medium. Same in an anisotropic medium.

2 2nd order nonlinearities

In a first step and for simplification, electromagnetic fields and macroscopic polarizations will be represented by scalar quantities. By doing so, it is assumed that the waves are polarized along the same direction, which will also be that of the macroscopic polarization. The second order nonlinear susceptibility will also be described by a frequency-independent scalar.

1. Assuming a non-vanishing 2nd order nonlinear response, give the expression of $\mathcal{P}^{(2)}(z,t)$, the 2nd order nonlinear polarization as a function of $\mathcal{E}(z,t)$. Following a spectral decomposition, identify the nonlinear effects that will be associated with each of the terms.
2. In general, two beams at ω_1 and ω_2 propagate in this medium, identify the set of effects that could occur during their propagation. Give an expression of the complex magnitude of the second order nonlinear polarization that is associated with each effect.

3 3rd order nonlinearities

As previously, electromagnetic fields and macroscopic polarizations will be represented by scalar quantities. The 3rd order nonlinear susceptibility will also be described by a frequency-independent scalar.

1. An ω beam propagates in a centro-symmetric medium (its nonlinear response at order 2 is zero), give the expression of the 3rd order nonlinear polarization, $\mathcal{P}^{(3)}(z,t)$.

2. What change in frequency can generate such an interaction?

3. Expressing the **<u>total</u>** macroscopic polarization $P(\omega)$ radiated at frequency ω. Show that the third-order nonlinearity modifies the linear susceptibility $\chi^{(1)}$ of the medium (*Subsidiary question: to which material parameters are the real and imaginary parts of the linear susceptibility related?*). How? What are the expected effects on beam propagation?

4. Same question as above but considering that a **weak** ω beam propagates with a **pump** beam ω_p of much higher intensity (we are still interested in the term $P(\omega)$ radiated at ω).

4 Physical origin of nonlinearities - Classical oscillator model

1. From the model of the harmonic oscillator, give the expression of the real and imaginary parts of the linear susceptibility $\chi^{(1)}(\omega)$ and recall to which properties of the material they are related. Comment on their variations with frequency.

2. What's the expression of $\chi^{(1)}(-\omega)$? Is it consistent with the complex notations used for the fields (see 1)?

3. What is the expression of $\chi^{(1)}(\omega)$ when the ω frequency is very small respectively to the resonant frequency associated with the medium (in the specific case of the model, a two-state medium)? When the frequency of a laser is very far from any transition, what simplifications can we make about the susceptibility of the medium?

4. What is the physical origin of nonlinearity in the electronic response of a dielectric medium?

5. In the expression $\chi^{(2)}(\omega_3,\omega_1,\omega_2)$ of the 2nd order nonlinear susceptibility, what do the frequency arguments refer to? What is their relationship?

6. In which situation, the frequency dispersion of the 2nd order nonlinear susceptibility can be neglected?

7. Using the oscillator motion equation (1.19) presented in the lecture notes, show that there is no non-zero 2nd order solution.

8. Show that the equation (1.19) is reduced to equation (1.20).

9. Solve this equation in the case of the interaction of two beams at the frequencies ω_1 and $\omega_2 = \omega_1 + \Omega$ and show that their interaction generates a polarization term at the frequency $\omega_3 = \omega_1 - \Omega$. Express the associated susceptibility. What are the expected effects of such an interaction?

10. The third-order nonlinear susceptibility also depends on the frequency $\chi^{(3)}(\omega_4;\omega_1,\omega_2,\omega_3)$. Give the relationship between the frequency arguments. In which situation, the frequency dispersion of the 3rd order nonlinear susceptibility can be neglected? On the contrary, in which situation(s) will this susceptibility be reinforced?