

Problem Set 4

Nonlinear Optics (NLO)

Due: 13. May 2015

1) Nonlinear polarization of n -th order

In Eq. (2.30) in the lecture notes we have used the following expansion for the electric field in the time domain:

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{2} \left(\sum_{l=-N}^N (1 + \delta_{l,0}) \underline{\mathbf{E}}(\mathbf{r}, \omega_l) e^{j\omega_l t} \right), \quad (1.1)$$

where $\delta_{j,k}$ is the Kronecker delta, i.e. $\delta_{j,k} = 0$ if $j \neq k$ and $\delta_{j,k} = 1$ if $j = k$, $\omega_{-l} = -\omega_l$, $\underline{\mathbf{E}}(\omega_l) = \underline{\mathbf{E}}^*(-\omega_l)$, $\omega_0 = 0$, and $\underline{\mathbf{E}}(\omega_0) \in \mathbb{R}$. Based on this relation, the complex time-domain amplitude of the n -th order polarization at a frequency $\omega_p = \omega_{l_1} + \dots + \omega_{l_n}$ can be written according to Eq. (2.32) in the lecture notes,

$$\underline{\mathbf{P}}^{(n)}(\omega_p) = \frac{1}{2^{n-1}} \epsilon_0 \sum_{\mathbb{S}(\omega_p)} \frac{(1 + \delta_{l_1,0}) \dots (1 + \delta_{l_n,0})}{1 + \delta_{p,0}} \underline{\chi}^{(n)}(\omega_p : \omega_{l_1}, \dots, \omega_{l_n}) : \underline{\mathbf{E}}(\omega_{l_1}) \dots \underline{\mathbf{E}}(\omega_{l_n}), \quad (1.2)$$

where $\mathbb{S}(\omega_p) = \{(l_1, \dots, l_n) | \omega_{l_1} + \dots + \omega_{l_n} = \omega_p\}$. Every frequency $\omega_{l_1}, \dots, \omega_{l_n}$ can take the positive or negative value of a frequency $\omega_1, \dots, \omega_n$ that appears in the input signal. The frequency-dependent susceptibility tensor $\underline{\chi}^{(n)}(\omega_p : \omega_{l_1}, \dots, \omega_{l_n})$ describes the nonlinear interaction between different electric field vectors.

1. Explain the meaning of the “:” sign in Eq. (1.2).
2. Apply Eq. (1.2) to the case of the nonlinear processes listed below and write down the complex time-domain amplitude $\underline{\mathbf{P}}^{(n)}$ of the nonlinear polarization as a function of the complex electric field amplitudes $\underline{\mathbf{E}}$. Sketch the energy-level diagrams involving all relevant virtual energy levels of the input frequencies.
 - a. Self-phase modulation (SPM): $\omega_p = \omega_1 + \omega_1 - \omega_1 = \omega_1$
 - b. Cross-phase modulation (XPM): $\omega_p = \omega_1 + \omega_2 - \omega_2 = \omega_1$
 - c. Non-degenerate four-wave mixing (non-deg. FWM): $\omega_p = \omega_1 + \omega_2 - \omega_3 = \omega_4$
 - d. Sum-frequency generation (SFG): $\omega_3 = \omega_1 + \omega_2$
 - e. Optical rectification (OR): $\omega_2 = \omega_1 - \omega_1$
 - f. Electro-optic Kerr effect: $\omega_3 = \omega_1 + \omega_2 + \omega_2 = \omega_1$, $\omega_2 = 0$

- For the case of SFG express the x -component of the complex time-domain amplitude of the nonlinear polarization $\underline{\mathbf{P}}^{(2)}(\omega_3 = \omega_1 + \omega_2)$. Consider the contributions of all vector components of the electric field and write down the fully expanded expression without using the tensorial short form notation.

2) Lorentz oscillator model for the linear case

In a classical picture, an electron bound to an atom can be considered as a harmonic oscillator in analogy to a mass connected to a spring. This oscillator, when driven by an external electric field E_x , follows the classical equation of motion

$$m_e \frac{d^2 x(t)}{dt^2} = -eE_x(t) - m_e \omega_r^2 x(t) - m_e \gamma_r \frac{dx(t)}{dt}, \quad (2.1)$$

where $x(t)$ is the dislocation of the electron, m_e denotes the electronic mass, $-eE_x(t)$ is the driving force by the external electric field, $-m_e \omega_r^2 x$ is the restoring force of the oscillator and $-m_e \gamma_r \frac{dx}{dt}$ is a damping term, with damping constant γ_r . The parameter ω_r will turn out to be the resonance frequency of the oscillator.

- Solve the differential equation (2.1) for a time-harmonic electric field of the form $E(t) = \frac{1}{2}(\underline{E}(\omega) \exp(j\omega t) + c.c.)$ by using a similar ansatz for the dislocation. Derive an expression for $\underline{x}(\omega)$.
- The electric polarization is the dipole moment per volume,

$$\underline{P}_x(\omega) = \varepsilon_0 \underline{\chi}^{(1)}(\omega) \underline{E}_x(\omega) = -\frac{N}{V} e \cdot \underline{x}(\omega), \quad (2.2)$$

where $\frac{N}{V}$ is the number density of atoms in the medium and $-e \cdot \underline{x}(\omega)$ is the induced dipole moment per atom. Show that the susceptibility is given by

$$\underline{\chi}^{(1)}(\omega) = \frac{Ne^2}{Vm_e \varepsilon_0} \frac{1}{\omega_r^2 - \omega^2 + j\omega_r \gamma_r} \quad (2.3)$$

and separate the susceptibility into real and imaginary part.

- Sketch the real and imaginary part of the susceptibility $\underline{\chi}^{(1)}$ around the resonance frequency ω_r . What is the consequence of this result for the real and imaginary part of the refractive index at very large, e.g. X-ray frequencies.

Questions and Comments:

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