Solution to Problem Set 4 Nonlinear Optics (NLO)

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:

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

where $\delta_{j,k}$ is the Kronecker delta, i.e. $\delta_{j,k} = 0$ for $j \neq k$ and $\delta_{j,k} = 1$ for 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 $\omega_p = \omega_{l_1} + ... + \omega_{l_n}$ can be written according to Eq. (2.32) in the lecture notes,

$$\underline{\mathbf{P}}^{(n)}(\boldsymbol{\omega}_{p}) = \frac{1}{2^{n-1}} \epsilon_{0} \sum_{\mathbb{S}(\boldsymbol{\omega}_{p})} \frac{\left(1 + \delta_{l_{1},0}\right) \dots \left(1 + \delta_{l_{n},0}\right)}{1 + \delta_{p,0}} \underline{\chi}^{(n)} \left(\boldsymbol{\omega}_{p} : \boldsymbol{\omega}_{l_{1}}, \dots, \boldsymbol{\omega}_{l_{n}}\right) \vdots \underline{\mathbf{E}}(\boldsymbol{\omega}_{l_{1}}) \dots \underline{\mathbf{E}}(\boldsymbol{\omega}_{l_{n}}), \quad (1.2)$$

where $\mathbb{S}(\omega_p) = \{(l_1, ..., l_n) | \omega_{l_1} + ... + \omega_{l_n} = \omega_p\}$. Every frequency $\omega_{l_1}, ..., \omega_{l_n}$ can take the positive or negative value of a frequency $\omega_1, ..., \omega_n$ that appears in the input signals. The frequency-dependent susceptibility tensor $\underline{\chi}^{(n)}(\omega_p : \omega_{l_1}, ..., \omega_{l_n})$ 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 of the nonlinear polarization as a function of the complex electric field amplitudes. Sketch the energy-level diagram involving all possible virtual electronic transitions 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: $\omega_3 = \omega_1 + \omega_2 + \omega_2 = \omega_1$, $\omega_2 = 0$
- 3. For the case of SFG express the *x*-component of the complex time-domain amplitude of the nonlinear polarization $\underline{\mathbf{P}}^{(2)}(\omega_3 = \omega_1 + \omega_2)$, without using the short form notation, i.e. using the tensor components $\underline{\chi}_{qr,s}^{(2)}$, where q = x.

Solution:

1. In the general case the short form tensor notation can be written as

$$\chi^{(n)} : \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \dots \mathbf{E}(\omega_n) = \sum_{q_0, q_1 \dots q_n} \mathbf{e}_{q_0} \chi^{(n)}_{q_0: q_1 q_2 \dots q_n} E_{q_1}(\omega_1) E_{q_2}(\omega_2) \dots E_{q_n}(\omega_n).$$
(1.3)

The " \vdots " sign therefore denotes the component-by-component multiplication and summation of a *n*-th rank tensor and *n* electric field vectors.

2.
a.
$$\mathbf{\underline{P}}^{(3)}(\omega_{1}) = \frac{3}{4} \epsilon_{0} \underline{\chi}^{(3)} (\omega_{1} : \omega_{1}, \omega_{1}, -\omega_{1}) : \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}^{*}(\omega_{1})$$
b.
$$\mathbf{\underline{P}}^{(3)}(\omega_{1}) = \frac{6}{4} \epsilon_{0} \underline{\chi}^{(3)} (\omega_{1} : \omega_{1}, \omega_{2}, -\omega_{2}) : \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}(\omega_{2}) \mathbf{\underline{E}}^{*}(\omega_{2})$$
c.
$$\mathbf{\underline{P}}^{(3)}(\omega_{4}) = \frac{6}{4} \epsilon_{0} \underline{\chi}^{(3)} (\omega_{4} : \omega_{1}, \omega_{2}, -\omega_{3}) : \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}(\omega_{2}) \mathbf{\underline{E}}^{*}(\omega_{3})$$
d.
$$\mathbf{\underline{P}}^{(2)}(\omega_{3}) = \epsilon_{0} \underline{\chi}^{(2)} (\omega_{3} : \omega_{1}, \omega_{2}) : \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}(\omega_{2})$$
e.
$$\mathbf{\underline{P}}^{(2)}(\omega_{2} = 0) = \frac{1}{2} \epsilon_{0} \underline{\chi}^{(2)} (0 : \omega_{1}, -\omega_{1}) : \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}^{*}(\omega_{1})$$
f.
$$\mathbf{\underline{P}}^{(3)}(\omega_{3}) = \frac{3}{4} \epsilon_{0} \underline{\chi}^{(3)} (\omega_{3} : \omega_{1}, \omega_{0}, \omega_{0}) : \mathbf{\underline{E}}(\omega_{1}) \mathbf{\underline{E}}(\omega_{0}) \mathbf{\underline{E}}(\omega_{0}) \mathbf{4}$$
J.
$$\mathbf{\underline{P}}^{(2)}(\omega_{3}) = \epsilon_{0} \sum_{r,s} \underline{\chi}^{(2)}_{s,r,s} \mathbf{\underline{E}}_{r}(\omega_{1}) \mathbf{\underline{E}}_{s}(\omega_{2})$$

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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

$$m_{\rm e} \frac{d^2 x(t)}{dt^2} = -eE_{\rm x}(t) - m_{\rm e} \omega_{\rm r}^2 x(t) - m_{\rm e} \gamma_{\rm r} \frac{dx(t)}{dt}, \qquad (2.1)$$

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. The parameter ω_r will turn out to be the resonance frequency of the oscillator.

- 1. 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 $x(\omega)$.
- 2. The electric polarization is the dipole moment per volume,

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

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

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

and separate the susceptibility into real and imaginary part.

3. 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.

Solution

1. We insert the ansatz for the electric field $E(t) = \frac{1}{2} (\underline{E}(\omega) \exp(j\omega t) + c.c.)$ and the dislocation $x(t) = \frac{1}{2} (\underline{x}(\omega) \exp(j\omega t) + c.c.)$ into the differential equation (2.1). We only take into account the terms oscillating at the same angular frequency and obtain:

$$-\omega^2 m_{\rm e} \underline{x}(\omega) = -e \underline{E}(\omega) - m_{\rm e} \omega_{\rm r}^2 \underline{x}(\omega) - j \omega m_{\rm e} \gamma_{\rm r} \underline{x}(\omega)$$

We solve for $\underline{x}(\omega)$ and get the frequency dependent dislocation of the oscillator:

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$$\underline{x}(\omega) = \frac{-e\underline{E}(\omega)}{m_{\rm e}(\omega_{\rm r}^2 - \omega^2 + j\omega\gamma_{\rm r})}$$

2. The polarization of the charged system (atom and electron) is given by $-e \cdot \underline{x}(\omega)$. As the total electric polarization of the medium is given by all of its atomic dipoles, we multiply with the number density $\frac{N}{V}$ of atoms in the medium. We then use Eq. (2.2) to relate the result to the 1st order susceptibility:

$$\underline{P}_{\mathbf{x}}(\omega) = \varepsilon_0 \underline{\chi}^{(1)}(\omega) \underline{E}_{\mathbf{x}}(\omega) = -\frac{N}{V} e \cdot \underline{x}(\omega) \Longrightarrow \underline{\chi}^{(1)}(\omega) = \frac{N e^2}{V m_{e} \varepsilon_0} \frac{1}{\omega_{\mathbf{r}}^2 - \omega^2 + j \omega_{\mathbf{r}} \gamma_{\mathbf{r}}}$$

We separate the susceptibility into real and imaginary part by expanding the fraction:

$$\begin{split} \underline{\chi}^{(1)}(\omega) &= \frac{Ne^2}{Vm_e\varepsilon_0} \frac{1}{\omega_r^2 - \omega^2 + j\omega_r\gamma_r} \cdot \frac{\omega_r^2 - \omega^2 - j\omega_r\gamma_r}{\omega_r^2 - \omega^2 - j\omega_r\gamma_r} = \frac{Ne^2}{Vm_e\varepsilon_0} \frac{\omega_r^2 - \omega^2 - j\omega_r\gamma_r}{\left(\omega_r^2 - \omega^2\right)^2 + \left(\omega_r\gamma_r\right)^2} \\ \chi^{(1)} &= \frac{Ne^2}{Vm_e\varepsilon_0} \frac{\omega_r^2 - \omega^2}{\left(\omega_r^2 - \omega^2\right)^2 + \left(\omega_r\gamma_r\right)^2} \\ \chi^{(1)}_i &= -\frac{Ne^2}{Vm_e\varepsilon_0} \frac{\omega_r\gamma_r}{\left(\omega_r^2 - \omega^2\right)^2 + \left(\omega_r\gamma_r\right)^2} \end{split}$$

3. Sketch of the susceptibility around the resonance frequency:





As derived in Problem Set 1, the real part of the refractive index $n \approx \sqrt{1 + \chi^{(1)}}$. Sketching this, it can be seen that the refractive index is smaller than one for frequencies above the resonance frequency (e.g. X-ray). Hence for building lenses for X-ray frequencies one has to use concave instead of convex structures.



Questions and Comments:

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