Solution to Problem Set 4
Nonlinear Optics (NLO)

1) Typical electric field strengths in nonlinear optics, typical nonlinear susceptibilities

For an order-of-magnitude estimation of the nonlinear susceptibilities $\chi^{(2)}$ and $\chi^{(3)}$, let us consider a simple model of a Hydrogen atom, in which the electron is bound to the nucleus by an electric field with the radial component that corresponds to a characteristic atomic field strength

$$E_{\text{at}} = \frac{e}{\epsilon_0 4\pi a_0^2}, \tag{1.1}$$

where $e = 1.60\times10^{-19}\,\text{C}$ is the elementary charge, $\epsilon_0 = 8.85\times10^{-12}\,\text{F/m}$ is the vacuum permittivity and $a_0 = 4\pi\epsilon_0\hbar^2/m_e e^2 = 0.053\,\text{nm}$ is the Bohr radius, i.e. the expectation value of the distance between the nucleus and the electron. The quantity $\hbar = 6.626\times10^{-34}\,\text{Js}$ denotes the reduced Planck constant, and $m_e = 9.1\times10^{-31}\,\text{kg}$ is the electron mass. Externally applied electric fields lead to a displacement of the electron with respect to the core and hence to an electric dipole moment, i.e., a non-zero electric polarization P. Usually external fields (E_{ext}) are much smaller than E_{at} and the dependence between E_{ext} and P can be approximated by the linear relationship $P = \epsilon_0\chi^{(1)}E_{\text{ext}}$. For most solid-state materials, $\chi^{(1)}$ is in the order of unity.

A common argument [1] says that the nonlinear contributions to the polarization $P_{\text{NL}}^{(m)} = \epsilon_0\chi^{(m)}E^m$ ($m > 1$) become comparable to the linear contribution $P_{\text{L}} = \epsilon_0\chi^{(1)}E$ when the applied electric field E_{ext} is of the order of E_{at}.

1. Calculate the characteristic atomic field strength E_{at}. Then, set $\chi^{(1)} = 1$ and assume for the radial component of the polarization that $P_{\text{L}} = P_{\text{NL}}^{(m)}$ for $E_{\text{ext}} = E_{\text{at}}$. Use these relations to estimate the order of magnitude of $\chi^{(2)}$ and $\chi^{(3)}$.

A typical femtosecond laser system produces pulses with a repetition rate of 80 MHz, a pulse duration of 100 fs, and an average power of 1 W at a wavelength of 1 µm. Imagine that the beam is focused on a spot having diameter equal to the wavelength.

2. What is the optical power averaged on one pulse, assuming that the pulse has rectangular shape?
3. What is the maximum electric field strength in the focus assuming that the field is uniform within the given diameter? Compare the magnitude of this field with E_{at}.

[1] Boyd, R.W., *Nonlinear Optics* (San Diego: Academic Press, 2003).

Solution:

1.

$$E_{\text{at}} = 5.13\times10^{11}\,\frac{\text{V}}{\text{m}}$$

$$\chi^{(2)} = \frac{1}{E_{\text{at}}} = 1.95\times10^{-12}\,\frac{\text{m}}{\text{V}}$$

$$\chi^{(3)} = \frac{1}{E_{\text{at}}^2} = 3.81\times10^{-24}\,\frac{\text{m}^2}{\text{V}^2}$$

2.

$$P_{\text{avg}} = 1\text{W}$$

$$P_{\text{pulse}} = \frac{12.5\text{ns}}{100\text{fs}}\cdot P_{\text{avg}} = 125\text{kW}$$

3.

$$I_{\text{pulse}} = \frac{P_{\text{pulse}}}{\pi\left(0.5\mu\text{m}\right)^2} = 1.59\times10^{17}\,\frac{\text{W}}{\text{m}^2}$$

$$E_{\text{pulse}} = \sqrt{\frac{2I_{\text{pulse}}}{c\epsilon_0}} = 1.09\times10^{10}\,\frac{\text{V}}{\text{m}}$$

where c is the speed of light in vacuum. This field is about a factor of 40 less than E_{at}, however this can be resolved with the sensitivity of today's equipment.

2) 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:

$$\mathbf{E}(\mathbf{r},t) = \frac{1}{2}\left(\sum_{l=-N}^{N}\left(1+\delta_{l,0}\right)\underline{\mathbf{E}}(\mathbf{r},\omega_l)e^{\mathrm{j}\omega_l t}\right), \tag{1.2}$$

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 $\omega_p=\omega_{l_1}+\ldots+\omega_{l_n}$ can be written according to Eq. (2.32) in the lecture notes,

$$\underline{\mathbf{P}}^{(n)}(\omega_p) = \frac{1}{2^{n-1}}\epsilon_0\sum_{\mathbb{S}(\omega_p)}\frac{\left(1+\delta_{l_1,0}\right)\ldots\left(1+\delta_{l_n,0}\right)}{1+\delta_{p,0}}\underline{\chi}^{(n)}\left(\omega_p:\omega_{l_1},\ldots,\omega_{l_n}\right)\vdots\underline{\mathbf{E}}(\omega_{l_1})\ldots\underline{\mathbf{E}}(\omega_{l_n}), \tag{1.3}$$

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

1. Explain the meaning of the "$\vdots$" sign in Eq. (1.3).
2. Apply Eq. (1.3) 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 $\underline{\chi}^{(2)}_{q:r,s}$, where $q=x$.

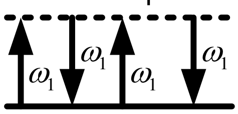
Solution:

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

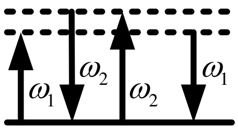
$$\chi^{(n)}\vdots\mathbf{E}\left(\omega_1\right)\mathbf{E}\left(\omega_2\right)\ldots\mathbf{E}\left(\omega_n\right)=\sum_{q_0,q_1\ldots,q_n}\mathbf{e}_{q_0}\chi^{(n)}_{q_0:q_1q_2\ldots q_n}E_{q_1}\left(\omega_1\right)E_{q_2}\left(\omega_2\right)\ldots E_{q_n}\left(\omega_n\right). \qquad (1.4)$$

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

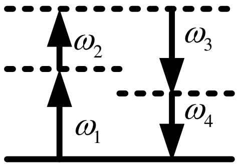
2.
    a. $\underline{\mathbf{P}}^{(3)}(\omega_1)=\frac{3}{4}\epsilon_0\underline{\chi}^{(3)}\left(\omega_1:\omega_1,\omega_1,-\omega_1\right)\vdots\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}^*(\omega_1)$



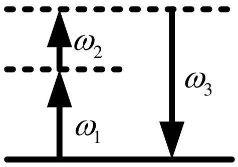
    b. $\underline{\mathbf{P}}^{(3)}(\omega_1)=\frac{6}{4}\epsilon_0\underline{\chi}^{(3)}\left(\omega_1:\omega_1,\omega_2,-\omega_2\right)\vdots\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}(\omega_2)\underline{\mathbf{E}}^*(\omega_2)$



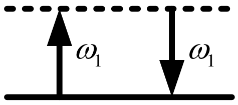
c. $\underline{\mathbf{P}}^{(3)}(\omega_4)=\frac{6}{4}\epsilon_0\underline{\chi}^{(3)}\left(\omega_4:\omega_1,\omega_2,-\omega_3\right):\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}(\omega_2)\underline{\mathbf{E}}^*(\omega_3)$



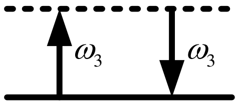
d. $\underline{\mathbf{P}}^{(2)}(\omega_3)=\epsilon_0\underline{\chi}^{(2)}\left(\omega_3:\omega_1,\omega_2\right):\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}(\omega_2)$



e. $\underline{\mathbf{P}}^{(2)}(\omega_2=0)=\frac{1}{2}\epsilon_0\underline{\chi}^{(2)}\left(0:\omega_1,-\omega_1\right):\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}^*(\omega_1)$



f. $\underline{\mathbf{P}}^{(3)}(\omega_3)=3\epsilon_0\underline{\chi}^{(3)}\left(\omega_3:\omega_1,\omega_0,\omega_0\right):\underline{\mathbf{E}}(\omega_1)\underline{\mathbf{E}}(\omega_0)\underline{\mathbf{E}}(\omega_0)$



3. $\underline{P}_x^{(2)}(\omega_3)=\epsilon_0\sum_{r,s}\underline{\chi}_{x:r,s}^{(2)}\underline{E}_r\left(\omega_1\right)\underline{E}_s\left(\omega_2\right)$

Questions and Comments:

Pablo Marin
Building: 30.10, Room: 2.33
Phone: 0721/608-42487
pablo.marin@kit.edu

Philipp Trocha
Room: 2.32/2
42480
philipp.trocha@kit.edu