University of Hamburg, Department of Physics

Nonlinear Optics

Kärtner/Mücke, WiSe 2018/2019

Problem Set 2

Issued: 02.11.2018 Due : 09.11.2018

1. **Coupled oscillator model for the nonlinear susceptibility - part 2.**

Do you remember our coupled oscillator model from the last problem set? So there we were using a simple model accounting for anharmonicity in the potentials for both ions and electrons via an anharmonic coupling term between the two. Now let's finally look into the nonlinear susceptibility and what we can learn from this model. Via mixing two optical frequencies ω_1 and ω_2 one can generate a field at frequency $\Omega = \omega_1 - \omega_2$ via difference frequency generation (DFG). In our case, we are interested in the process of THz generation meaning that we choose the frequencies such that Ω is in the THz range.

Your task now will be to derive an expression for the frequency dependent $\chi^{(2)}(\Omega; \omega_1, -\omega_2)$ susceptibility (which is one of the several contributions to the total $\chi^{(2)}$ tensor) within our model. To do so, please refer back to problem 2 on the last problem set (you may also take a look at the solution, in case you didn't get as far) to now solve the equation of motion for the next perturbation term $x^{(2)}$ (see equations (2) and (4)). To guide you through this problem, these are the steps you may take (but feel free to find a different solution yourself):

$$\ddot{x_i}^{(1)} + \omega_i^2 x_i^{(1)} + \gamma_i \dot{x_i}^{(1)} = \frac{q_i E(t)}{m_i} \tag{1}$$

$$\ddot{x_i}^{(2)} + \omega_i^2 x_i^{(2)} + \gamma_i \dot{x_i}^{(2)} = \frac{\beta}{m_i}((x_i^{(1)})^2 + (x_e^{(1)})^2 - 2x_e^{(1)} x_i^{(1)}) \tag{2}$$

$$\ddot{x_e}^{(1)} + \omega_e^2 x_e^{(1)} + \gamma_e \dot{x_e}^{(1)} = \frac{q_e E(t)}{m_e} \tag{3}$$

$$\ddot{x_e}^{(2)} + \omega_e^2 x_e^{(2)} + \gamma_e \dot{x_e}^{(2)} = \frac{\beta}{m_e}((x_e^{(1)})^2 + (x_i^{(1)})^2 - 2x_i^{(1)} x_e^{(1)}) \tag{4}$$

a) Since we want to obtain $\chi^{(2)}(\Omega; \omega_1, -\omega_2)$, we need to first get $x^{(2)}(\Omega = \omega_1 - \omega_2) = x_i^{(2)}(\Omega) + x_e^{(2)}(\Omega)$ via solving equations (2) and (4). Let's start by looking at their driving terms composed of $x_i^{(1)} x_i^{(1)} + x_e^{(1)} x_i^{(1)} + x_e^{(1)} x_e^{(1)}$, which are the solutions obtained for the first order equations. As we are looking into

DFG of two frequencies ω_1 and ω_2, we now have to consider a driving electric field $E(t) = E_1(t) + E_2(t)$ of the form:

$$\begin{aligned} E(t) = \left(E_0 e^{i\omega_1 t} + E_0 e^{-i\omega_1 t}\right) + \left(E_0 e^{i\omega_2 t} + E_0 e^{-i\omega_2 t}\right) \\ = E_0 e^{i\omega_1 t} + E_0 e^{i\omega_2 t} + c.c. \end{aligned}$$

As equations (1) and (3) are **linear** differential equations, we can easily obtain their solutions $x_i(\omega_1, \omega_2)$ and $x_e(\omega_1, \omega_2)$for the new field composed of two frequencies via the sum of solutions for just a single frequency:

$$x_i(\omega_1, \omega_2) = \frac{q_i}{m_i} \frac{E_0 e^{i\omega_1 t}}{D_i(\omega_1)} + \frac{q_i}{m_i} \frac{E_0 e^{i\omega_2 t}}{D_i(\omega_2)} + c.c., \quad \text{with } D_i(\omega) = \omega_i^2 - \omega^2 + i\gamma_i \omega \quad (5)$$

$$x_e(\omega_1, \omega_2) = \frac{q_e}{m_e} \frac{E_0 e^{i\omega_1 t}}{D_e(\omega_1)} + \frac{q_e}{m_e} \frac{E_0 e^{i\omega_2 t}}{D_e(\omega_2)} + c.c., \quad \text{with } D_e(\omega) = \omega_e^2 - \omega^2 + i\gamma_e \omega \quad (6)$$

Using equations (5) and (6) now write down the explicit expressions for $x_i^{(1)} x_i^{(1)}$, $x_e^{(1)} x_i^{(1)}$ and $x_e^{(1)} x_e^{(1)}$!

b) Since we are only interested in the DFG process $x^{(2)}(\Omega)$, we only need to look at those terms in the driving term that will actually lead to a nonlinear polarization at with frequency $\Omega = \omega_1 - \omega_2$. Thus, neglect the other contributions (which give rise to sum frequency generation, frequency doubling and optical rectification,...) and rewrite $x_i^{(1)} x_i^{(1)}$, $x_e^{(1)} x_i^{(1)}$ and $x_e^{(1)} x_e^{(1)}$ only in terms of the contribution for difference frequency generation. Now proof that $x_e^{(1)} x_i^{(1)} \approx x_i^{(1)} x_i^{(1)} \approx 0$ (for this purpose your insights from the last problem set may come in handy).

c) So we have shown the driving term to be considered is given by:

$$(x_e^{(1)})^2(\Omega) = \left(\frac{q_e}{me}\right)^2 \frac{E_0^2 e^{i(\omega_1 - \omega_2)t}}{D_e(\omega_1) D_e(\omega_2)^*} = x_e^{(1)}(\omega_1) x_e^{(1)}(\omega_2)^* \quad (7)$$

Now we can finally solve equations (2) and (4). Looking at them, we find them to be of the same form as the first order equation but with a different driving term. Thus you can use the comparison to the first equation to come up with the solution for these two equations. Find the expressions for $x_i^{(2)}$ and $x_e^{(2)}$!

e) Now find the expression for $\chi^{(2)}(\Omega; \omega_1, -\omega_2) = \chi_i^{(2)}(\Omega; \omega_1, -\omega_2) + \chi_e^{(2)}(\Omega; \omega_1, -\omega_2)$ and then show that this can be rewritten in terms of the linear susceptibilities such as:

$$\chi^{(2)}(\Omega;\omega_1,-\omega_2) = \frac{\beta\epsilon_0^2}{N_i N_e q_i^2 q_e}\chi_i(\Omega)\chi_e(\omega_1)\chi_e(-\omega_2) + \frac{\beta\epsilon_0^2}{N_e^2 q_e^3}\chi_e(\Omega)\chi_e(\omega_1)\chi_e(-\omega_2) \tag{8}$$

f) Which contribution ($\chi_i^{(2)}$ or $\chi_e^{(2)}$) is the most crucial for THz generation within our model? Using a plotting program of your choice and the parameters given on the last problem set, please 3D plot or contour-plot the second order susceptibility over the frequency range of $0-4$ THz for Ω and $1100-1300$ THz for ω_1. Explain what Kleinman symmetry means and whether it holds for our model as well!

2. **Second order process in a PPLN crystal**

   Periodically poled $LiNbO_3$ (PPLN) is used to achieve quasi-phase matching (QPM) for a nonlinear process of second order. We want to describe a so called parametric process, frequently utilized in modern optics technology to generate or amplify selected frequencies. The three beams involved in such second order process are the so called signal, pump and idler beam. The crystal is pumped at $\lambda_p = 532nm$ and generates a signal beam at $\lambda_s = 950nm$.

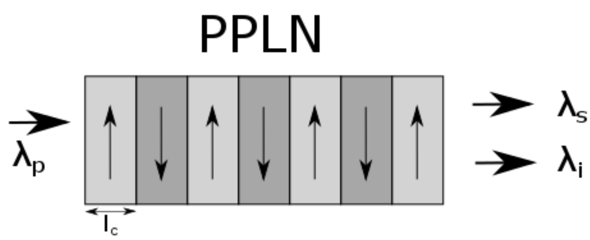

Figure 1: Sketch of the parametric process in a PPLN crystal.

   (a) Find the relations of frequency and wavelength between the three involved waves (pump, signal and idler) and determine the wavelength of the idler beam.

   (b) What component of the polarization should be used if we want to make use of the high nonlinear coefficient $d_{33}/\chi^{(2)}_{zzz}$ Write down the full expression for that component of the polarization.

(c) Evaluate the phase mismatch between the three waves in the $LiNbO_3$ crystal. You can look up the refractive indices at http://refractiveindex.info/. Remember what polarization we preferred from (b) when choosing ordinary or extraordinary refractive index. Calculate the coherence length $l_c = \frac{\pi}{\Delta k}$ for the given setup.