Solution to Problem Set 5
Nonlinear Optics (NLO)

1) Perturbative analysis of anharmonic oscillator for the nonlinear case

We want to expand the Lorentz oscillator model of the previous problem set (No. 2) to the nonlinear case. To this end, we assume that the electrons bound to the nucleus are subject to an anharmonic potential. In a simplified 1D-model we consider only a linear displacement along the radial direction, which, without loss of generality, shall be associated with the x coordinate. The potential can be written as

$$V(x) = \frac{1}{2} m_\mathrm{e} \omega_\mathrm{r}^2 x^2 + \frac{1}{3} m_\mathrm{e} \beta_2 x^3 + \frac{1}{4} m_\mathrm{e} \beta_3 x^4 \,,$$

where β_2 and β_3 are the parameters defining the strength of the anharmonicity. The force exerted on the electron by this potential is

$$F(x) = -\frac{\mathrm{d}V(x)}{\mathrm{d}x} = -m_\mathrm{e} \omega_\mathrm{r}^2 x - m_\mathrm{e} \beta_2 x^2 - m_\mathrm{e} \beta_3 x^3 \,.$$

For weak driving forces $F_d = -eE_\mathrm{x}(t)$ and small displacements we can assume the system to be only weakly anharmonic, i.e., $|\beta_2 x| + |\beta_3 x^2| << \omega_r^2$. We can then solve this problem by introducing a perturbation parameter λ into the solution ansatz for the displacement $x(t)$ of the center of the electron cloud from the nucleus,

$$x(t) = x_0(t) + \lambda x_1(t) + \lambda^2 x_2(t) + \ldots \,,$$

$$V(x) = \frac{1}{2} m_\mathrm{e} \omega_\mathrm{r}^2 x^2 + \lambda \frac{1}{3} m_\mathrm{e} \beta_2 x^3 + \lambda \frac{1}{4} m_\mathrm{e} \beta_3 x^4 \,.$$

In these relations $x_0(t)$ is the displacement for the case of the unperturbed harmonic oscillator. The deviation from the harmonic case is taken into account by a series of correction terms with higher orders of the perturbation parameter λ, where $\lambda \neq 0$ turns the anharmonicity on and $\lambda = 0$ turns it off. We adapt the equation of motion of the electron to the new case:

$$m_\mathrm{e} \frac{\mathrm{d}^2 x(t)}{\mathrm{d}t^2} = -eE_\mathrm{x}(t) - m_\mathrm{e} \omega_\mathrm{r}^2 x(t) - \lambda m_\mathrm{e} \beta_2 x^2(t) - \lambda m_\mathrm{e} \beta_3 x^3(t) - m_\mathrm{e} \gamma_\mathrm{r} \frac{\mathrm{d}x(t)}{\mathrm{d}t} \qquad (1.1)$$

1. Insert the ansatz for $x(t)$ up to the first order of λ into the differential Eq. (1.1) and find expressions for $\frac{\mathrm{d}^2 x_0(t)}{\mathrm{d}t^2}$, $\frac{\mathrm{d}^2 x_1(t)}{\mathrm{d}t^2}$ by comparing the coefficients associated with the various orders of the perturbation parameter λ. Show that the solutions are given by:

(0th order): $m_e \frac{d^2 x_0(t)}{dt^2} = -eE_x(t) - m_e \omega_r^2 x_0(t) - m_e \gamma_r \frac{dx_0(t)}{dt}$

(1st order): $m_e \frac{d^2 x_1(t)}{dt^2} = -m_e \omega_r^2 x_1(t) - m_e \gamma_r \frac{dx_1(t)}{dt} - m_e \beta_2 x_0^2(t) - m_e \beta_3 x_0^3(t)$

Solution

By inserting $x(t) = x_0(t) + \lambda x_1(t)$ into Eq. (1.1) we obtain:

$$\begin{aligned} m_e \frac{d^2 x_0(t)}{dt^2} + \lambda m_e \frac{d^2 x_1(t)}{dt^2} &= -eE_x(t) - m_e \omega_r^2 x_0(t) - \lambda m_e \omega_r^2 x_1(t) - \\ &-\lambda m_e \beta_2 x_0^2(t) + 2\lambda^2 m_e \beta_2 x_0(t) x_1(t) - \lambda^3 m_e \beta_2 x_1(t) - \\ &-m_e \beta_3 \left(\lambda x_0^3(t) - \cancel{3\lambda^2 x_0^2(t) x_1^2(t)} + \cancel{3\lambda^3 x_0(t) x_1(t)} - \cancel{\lambda^4 x_1^3(t)} \right) - \\ &-m_e \gamma_r \frac{dx_0(t)}{dt} - \lambda m_e \gamma_r \frac{dx_1(t)}{dt}. \end{aligned} \tag{1.2}$$

Since we have weak perturbation (weakly anharmonic system $\Rightarrow \lambda \to 0$), we can neglect all terms associated with $\lambda^2, \lambda^3, ...$ as indicated in Eq. (1.2). We can now extract the terms associated with λ^0 (0th order) and λ^1 (1st order):

(0th order): $$\frac{d^2 x_0(t)}{dt^2} = -\frac{e}{m_e} E_x(t) - \omega_r^2 x_0(t) - \gamma_r \frac{dx_0(t)}{dt} \tag{1.3}$$

(1st order): $$\frac{d^2 x_1(t)}{dt^2} = -\omega_r^2 x_1(t) - \gamma_r \frac{dx_1(t)}{dt} - \beta_2 x_0^2(t) - \beta_3 x_0^3(t) \tag{1.4}$$

The 0th order represents the case of a linear harmonic oscillator that was solved in the problem set 2 by using the ansatz $x_0(t) = \frac{1}{2}\left(\underline{x}(\omega) \exp(j\omega t) + c.c. \right)$.

2. For the 0th order, we already know the solution; it is the unperturbed harmonic oscillator. For the 1st order, explicitly take into account the solutions that oscillate at the fundamental, the second and third harmonic, and at DC, i.e., use the ansatz

$$\begin{aligned} x_1(t) = &\underline{x}_1(\omega = 0) + \\ &\frac{1}{2}\left(\underline{x}_1(\omega) \exp(j\omega t) + c.c. \right) + \\ &\frac{1}{2}\left(\underline{x}_1(2\omega) \exp(j2\omega t) + c.c. \right) + \\ &\frac{1}{2}\left(\underline{x}_1(3\omega) \exp(j3\omega t) + c.c. \right). \end{aligned}$$

Find the amplitudes $\underline{x}_1(\omega=0)$, $\underline{x}_1(\omega)$, $\underline{x}_1(2\omega)$, $\underline{x}_1(3\omega)$ by inserting the ansatz for $x_1(t)$ into the differential equation for the first order perturbation in λ and collecting terms oscillating at the same frequency.

Solution

The differential equation (1.4) connects the perturbative solution $x_1(t)$ with the solution of the unperturbed oscillator $x_0(t)$. We start by finding expressions for $x_0^2(t)$ and $x_0^3(t)$, and we also have to take into account the complex conjugate in the calculations:

$$x_0^2(t)=\frac{1}{4}\left(\underline{x}(\omega)e^{j\omega t}+c.c.\right)^2=\frac{1}{2}\left|\underline{x}_0(\omega)\right|^2+\frac{1}{4}\left(\underline{x}_0(\omega)^2 e^{j2\omega t}+c.c.\right) \tag{2.1}$$

$$x_0^3(t)=\frac{1}{8}\left(\underline{x}_0(\omega)^3 e^{j3\omega t}+3\left|\underline{x}_0(\omega)\right|^2\underline{x}_0(\omega)e^{j\omega t}+c.c.\right). \tag{2.2}$$

After inserting the ansatz for $x_1(t)$, as well as Eqs. (2.1) and (2.2) into Eq. (1.4), we get:

$$\begin{aligned}
&0-\frac{\underline{x}_1(\omega)}{2}\omega^2e^{j\omega t}-\frac{\underline{x}_1(2\omega)}{2}4\omega^2e^{j2\omega t}-\frac{\underline{x}_1(3\omega)}{2}9\omega^2e^{j3\omega t}=\\
&=-\omega_r^2\underline{x}_1(\omega=0)-\omega_r^2\frac{\underline{x}_1(\omega)}{2}e^{j\omega t}-\omega_r^2\frac{\underline{x}_1(2\omega)}{2}e^{j2\omega t}-\omega_r^2\frac{\underline{x}_1(3\omega)}{2}e^{j3\omega t}-\\
&\quad-\gamma_r j\omega\frac{\underline{x}_1(\omega)}{2}e^{j\omega t}-\gamma_r j2\omega\frac{\underline{x}_1(2\omega)}{2}e^{j2\omega t}-\gamma_r j3\omega\frac{\underline{x}_1(3\omega)}{2}e^{j3\omega t}-\\
&\quad-\frac{\beta_2}{2}\left|\underline{x}_0(\omega)\right|^2-\frac{\beta_2}{4}\underline{x}_0^2(\omega)e^{j2\omega t}-\frac{\beta_3}{8}\underline{x}_0^3(\omega)e^{j3\omega t}-\frac{3\beta_3}{8}\left|\underline{x}_0(\omega)\right|^2\underline{x}_0(\omega)e^{j\omega t}.
\end{aligned} \tag{2.3}$$

From Eq. (2.3), we can now extract the terms associated with their corresponding frequencies (0, ω, 2ω, and 3ω), and get the field amplitudes. By additionally using the following two relations (refer to the problem set 2):

$$\underline{x}_0(\omega)=-\frac{V\varepsilon_0}{Ne}\underline{E}(\omega)\underline{\chi}^{(1)}(\omega) \tag{2.4}$$

$$\underline{\chi}^{(1)}(\omega)\frac{Vm_\mathrm{e}\varepsilon_0}{Ne^2}=\frac{1}{\omega_r^2-\omega^2+j\omega\gamma_\mathrm{r}}, \tag{2.5}$$

we can reformulate the expressions for the field amplitudes:

$$\underline{x}_1(\omega=0)=-\frac{1}{2}\frac{\beta_2}{\omega_\mathrm{r}^2}\left|\underline{x}_0(\omega)\right|^2=-\frac{1}{2}\frac{\beta_2}{\omega_\mathrm{r}^2}\left(\frac{V\varepsilon_0}{Ne}\right)^2\left|\underline{E}(\omega)\right|^2\left|\underline{\chi}^{(1)}(\omega)\right|^2 \tag{2.6}$$

$$\begin{aligned}\underline{x}_1(\omega) &= -\frac{3}{4}\beta_3 \frac{\left|\underline{x}_0(\omega)\right|^2 \underline{x}_0(\omega)}{\omega_\mathrm{r}^2-\omega^2+j\omega\gamma_\mathrm{r}} = \\ &= -\frac{3}{4}\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^4 \frac{m_\mathrm{e}}{e}\underline{E}(\omega)\left|\underline{E}(\omega)\right|^2\left|\underline{\chi}^{(1)}(\omega)\right|^2\left(\underline{\chi}^{(1)}(\omega)\right)^2\end{aligned} \quad (2.7)$$

$$\begin{aligned}\underline{x}_1(2\omega) &= -\frac{1}{2}\beta_2 \frac{\underline{x}_0^2(\omega)}{\omega_\mathrm{r}^2-(2\omega)^2+j2\omega\gamma_\mathrm{r}} = \\ &= -\frac{1}{2}\beta_2\left(\frac{V\varepsilon_0}{Ne}\right)^3 \frac{m_\mathrm{e}}{e}\left(\underline{E}(\omega)\right)^2\underline{\chi}^{(1)}(2\omega)\left(\chi^{(1)}(\omega)\right)^2\end{aligned} \quad (2.8)$$

$$\begin{aligned}\underline{x}_1(3\omega) &= -\frac{1}{4}\beta_3 \frac{\underline{x}_0^3(\omega)}{\omega_\mathrm{r}^2-(3\omega)^2+j3\omega\gamma_\mathrm{r}} = \\ &= -\frac{1}{4}\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^4 \frac{m}{e}\left(\underline{E}(\omega)\right)^3\left(\underline{\chi}^{(1)}(\omega)\right)^3\underline{\chi}^{(1)}(3\omega)\end{aligned} . \quad (2.9)$$

Note that the 1st order susceptibilities at 2ω and 3ω appear in the expressions for $\underline{x}_1(2\omega)$ and $\underline{x}_1(3\omega)$.

3. Using the results from part 2, write down the electric polarizations $P(\omega_p)$ oscillating at the various frequencies ω_p. The polarizations can be related to the amplitudes of the displacement of the electron cloud by $P(\omega_p) = -\frac{N}{V}e\underline{x}_1(\omega_p)$, where $\frac{N}{V}$ is the number density of atoms in the medium and where $-e\cdot\underline{x}_1(\omega_p)$ is the induced nonlinear dipole moment per atom at the respective frequency ω_p.

Solution

After solving part 2, finding the electric polarization is trivial:

$$P(\omega=0) = \frac{Ne}{V}\frac{\beta_2}{2}\left(\frac{V\varepsilon_0}{Ne}\right)^3\left|\underline{E}(\omega)\right|^2\left|\underline{\chi}^{(1)}(\omega)\right|^2\underline{\chi}^{(1)}(0) \quad (3.1)$$

$$P(\omega) = -\frac{Ne}{V}\frac{3}{4}\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^4\frac{m_\mathrm{e}}{e}\underline{E}(\omega)\left|\underline{E}(\omega)\right|^2\left|\underline{\chi}^{(1)}(\omega)\right|^2\left(\underline{\chi}^{(1)}(\omega)\right)^2 \quad (3.2)$$

$$P(2\omega) = \frac{Ne}{V}\frac{1}{2}\beta_2\left(\frac{V\varepsilon_0}{Ne}\right)^3\frac{m_\mathrm{e}}{e}\left(\underline{E}(\omega)\right)^2\underline{\chi}^{(1)}(2\omega)\left(\chi^{(1)}(\omega)\right)^2 \quad (3.3)$$

$$P(3\omega) = -\frac{Ne}{V}\frac{1}{4}\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^4\frac{m}{e}\left(\underline{E}(\omega)\right)^3\left(\underline{\chi}^{(1)}(\omega)\right)^3\underline{\chi}^{(1)}(3\omega) . \quad (3.4)$$

4. Write down the general expressions for the nonlinear polarization in the scalar approximation for the following cases:
   a. Optical rectification
   b. Second harmonic generation
   c. Self-phase modulation
   d. Third harmonic generation

   Use the results from part 3. to derive expressions for the nonlinear susceptibilities:
   a. $\chi^{(2)}(0:\omega,-\omega)$
   b. $\chi^{(2)}(2\omega:\omega,\omega)$
   c. $\chi^{(3)}(\omega:\omega,\omega,-\omega)$
   d. $\chi^{(3)}(3\omega:\omega,\omega,\omega)$

Solution

The general expressions for the nonlinear polarization for the four cases are:

a. Optical rectification: $P=\frac{1}{2}\varepsilon_0\chi^{(2)}(0:\omega,-\omega)\underline{E}(\omega)\underline{E}^*(\omega)$ (4.1)

b. Second harmonic generation: $P=\frac{1}{2}\varepsilon_0\chi^{(2)}(2\omega:\omega,\omega)\underline{E}(\omega)\underline{E}(\omega)$ (4.2)

c. Self-phase modulation: $P=\frac{3}{4}\varepsilon_0\chi^{(3)}(\omega:\omega,\omega,-\omega)\underline{E}(\omega)\underline{E}(\omega)\underline{E}^*(\omega)$ (4.3)

d. Third harmonic generation: $P=\frac{1}{4}\varepsilon_0\chi^{(3)}(\omega:\omega,\omega,\omega)\underline{E}(\omega)\underline{E}(\omega)\underline{E}(\omega)$ (4.4)

In order to find the nonlinear susceptibilities, we need to compare the Eqs. (4.1) - (4.4) to the result from part 3 (Eqs. (3.1) - (3.4)).

a. $$\cancel{\frac{1}{2}}\varepsilon_0\chi^{(2)}(0:\omega,-\omega)\cancel{\underline{E}(\omega)\underline{E}^*(\omega)}=\frac{Ne}{V}\cancel{\frac{1}{2}}\frac{\beta_2}{\omega_\mathrm{r}^2}\left(\frac{V\varepsilon_0}{Ne}\right)^2\cancel{\left|\underline{E}(\omega)\right|^2}\left|\underline{\chi}^{(1)}(\omega)\right|^2$$

$$\chi^{(2)}(0:\omega,-\omega)=\frac{N\cancel{e}}{\cancel{V}\varepsilon_0}\frac{\beta_2}{\omega_\mathrm{r}^2}\left(\frac{V\varepsilon_0}{Ne}\right)^{\cancel{2}}\left|\underline{\chi}^{(1)}(\omega)\right|^2=\frac{\beta_2}{\omega_\mathrm{r}^2}\left(\frac{V\varepsilon_0}{Ne}\right)\left|\underline{\chi}^{(1)}(\omega)\right|^2$$

From Eq. (2.5) it follows: $\underline{\chi}^{(1)}(\omega=0)\frac{Vm_\mathrm{e}\varepsilon_0}{Ne^2}=\frac{1}{\omega_r^2}$, and if we substitute this in the last expression for $\chi^{(2)}(0:\omega,-\omega)$, we get:

$$\chi^{(2)}(0:\omega,-\omega)=\beta_2\frac{m_\mathrm{e}}{e}\left(\frac{V\varepsilon_0}{Ne}\right)^2\underline{\chi}^{(1)}(\omega=0)\left|\underline{\chi}^{(1)}(\omega)\right|^2 \tag{4.5}$$

In a similar fashion, we can find the expressions for the remaining three susceptibilities:

b. $$\chi^{(2)}\left(2\omega:\omega,\omega\right)=\beta_2\frac{m_\mathrm{e}}{e}\left(\frac{V\varepsilon_0}{Ne}\right)^2\underline{\chi}^{(1)}\left(2\omega\right)\left(\underline{\chi}^{(1)}\left(\omega\right)\right)^2 \tag{4.6}$$

c. $$\chi^{(3)}\left(\omega:\omega,\omega,-\omega\right)=\beta_3\frac{m_\mathrm{e}}{e}\left(\frac{V\varepsilon_0}{Ne}\right)^3\underline{\chi}^{(1)}\left(\omega\right)\left|\underline{\chi}^{(1)}\left(\omega\right)\right|^2 \tag{4.7}$$

d. $$\chi^{(3)}\left(3\omega:\omega,\omega,\omega\right)=\beta_3\frac{m}{e}\left(\frac{V\varepsilon_0}{Ne}\right)^3\underline{\chi}^{(1)}\left(3\omega\right)\left(\underline{\chi}^{(1)}\left(\omega\right)\right)^3. \tag{4.8}$$

Questions and Comments:

Pablo Marin-Palomo
Building: 30.10, Room: 2.33
Phone: 0721/608-42487

Philipp Trocha
Building: 30.10, Room: 2.32-2
Phone: 0721/608-42480

nlo@ipq.kit.edu