Problem Set 5
Nonlinear Optics (NLO)

Due: 9 June 2015

1) Perturbative analysis of anharmonic oscillator for the nonlinear case

We want to expand the Lorentz oscillator model of the previous problem set (No. 4) 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

$$V(x) = \frac{1}{2} m_e \omega_r^2 x^2 + \frac{1}{3} m_e \beta_2 x^3 + \frac{1}{4} m_e \beta_3 x^4 ,$$

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

$$F(x) = -\frac{\mathrm{d}V(x)}{\mathrm{d}x} = -m_e \omega_r^2 x - m_e \beta_2 x^2 - m_e \beta_3 x^3 .$$

For weak driving forces $F_d = -eE_x(t)$ and small displacements we can assume the system to be only weakly anharmonic, i.e., $\left|\beta_2 x + \beta_3 x^2\right| \leq \omega_r^2$. 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,

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

$$V(x) = \frac{1}{2} m_e \omega_r^2 x^2 + \lambda \frac{1}{3} m_e \beta_2 x^3 + \lambda \frac{1}{4} m_e \beta_3 x^4 .$$

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 $\lambda \to 1$ turns the anharmonicity on and $\lambda \to 0$ turns it off. We adapt our previous differential equation to the new case:

$$m_e \frac{\mathrm{d}^2 x(t)}{\mathrm{d}t^2} = -eE_x(t) - m_e \omega_r^2 x(t) - \lambda m_e \beta_2 x(t)^2 - \lambda m_e \beta_3 x(t)^3 - m_e \gamma_r \frac{\mathrm{d}x(t)}{\mathrm{d}t} \qquad (0.1)$$

1. Insert the ansatz for $x(t)$ up to the first order of λ into the differential Eq. (0.1) 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:

$$(0^{\text{th}}\text{ order}):\ m_{\mathrm{e}}\frac{\mathrm{d}^2 x_0(t)}{\mathrm{d}t^2} = -eE_{\mathrm{x}}(t) - m_{\mathrm{e}}\omega_{\mathrm{r}}^2 x_0(t) - m_{\mathrm{e}}\gamma_{\mathrm{r}}\frac{\mathrm{d}x_0(t)}{\mathrm{d}t}$$

$$(1^{\text{st}}\text{ order}):\ m_{\mathrm{e}}\frac{\mathrm{d}^2 x_1(t)}{\mathrm{d}t^2} = -m_{\mathrm{e}}\omega_{\mathrm{r}}^2 x_1(t) - m_{\mathrm{e}}\gamma_{\mathrm{r}}\frac{\mathrm{d}x_1(t)}{\mathrm{d}t} - m_{\mathrm{e}}\beta_2 x_0(t)^2 - m_{\mathrm{e}}\beta_3 x_0(t)^3$$

2. For the 0^{th} order, we already know the solution - it is the unperturbed harmonic oscillator. For the 1^{st} order, explicitly take into account the solutions that oscillate at the fundamental, the second and third harmonic, or 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!

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.

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

1. We insert $x(t)=x_0(t)+\lambda x_1(t)$ into equation (0.1) and obtain:

$$\frac{d^2\left(x_0+\lambda x_1\right)}{dt^2}=-\frac{e}{m_\mathrm{e}}E_\mathrm{x}-\omega_\mathrm{r}^2\left(x_0+\lambda x_1\right)-\lambda\beta_2\left(x_0+\lambda x_1\right)^2-\lambda\beta_3\left(x_0+\lambda x_1\right)^3-\gamma_\mathrm{r}\frac{d\left(x_0+\lambda x_1\right)}{dt}$$

$$\frac{d^2x_0}{dt^2}+\frac{\lambda d^2x_1}{dt^2}=-\frac{e}{m_\mathrm{e}}E_\mathrm{x}-\omega_\mathrm{r}^2x_0-\lambda\omega_\mathrm{r}^2x_1-\lambda\beta_2x_0^2-\lambda\beta_3x_0^3+ \tag{0.2}$$

$$-\gamma_\mathrm{r}\frac{dx_0}{dt}-\lambda\gamma_\mathrm{r}\frac{dx_1}{dt}+\mathcal{O}(\lambda^2)+\mathcal{O}(\lambda^3)+\mathcal{O}(\lambda^4)$$

We stop calculating terms larger than 1st order in λ on the right hand side as these terms do not find a counterpart on the left hand side. The terms with the same order in λ on both sides are equated:

(0th order): $$\frac{d^2x_0(t)}{dt^2}=-\frac{e}{m_\mathrm{e}}E_\mathrm{x}(t)-\omega_\mathrm{r}^2x_0(t)-\gamma_\mathrm{r}\frac{dx_0(t)}{dt} \tag{0.3}$$

(1st order): $$\frac{d^2x_1(t)}{dt^2}=-\omega_\mathrm{r}^2x_1(t)-\gamma_\mathrm{r}\frac{dx_1(t)}{dt}-\beta_2x_0(t)^2-\beta_3x_0(t)^3 \tag{0.4}$$

The 0th order represents the case of a linear harmonic oscillator which was solved in the problem set 4 by using an ansatz $x_0(t)=\frac{1}{2}\left(\underline{x}(\omega)\exp(j\omega t)+c.c.\right)$.

2. The differential equation (0.4) connects the perturbative solution $x_1(t)$ with the solution of the unperturbed oscillator $x_0(t)$. To simplify things, we start by finding expressions for $x_0(t)^2$ and $x_0(t)^3$, where we also have to take into account the complex conjugate in the calculations:

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

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

We expect to obtain nonlinearities (DC, fundamental, 2nd and 3rd harmonic), therefore we choose a solution that already contains them. We go on by inserting the given ansatz for $x_1(t)$ into differential equation (0.4) and can right away equate the terms oscillating at the same frequency. As $\underline{x}_0(\omega)$ will be present in the solution, we can express the nonlinear dislocations by the first order susceptibility.

(**Reminder:** $\underline{x}_0(\omega) = -\frac{V\varepsilon_0}{Ne}\underline{E}\underline{\chi}^{(1)}(\omega)$ and $\underline{\chi}^{(1)}(\omega)\frac{Vm_e\varepsilon_0}{Ne^2} = \frac{1}{\omega_r - \omega + j\omega\gamma_r}$)

- Terms at $\omega = 0$ give us: $0 = -\omega_r^2 \underline{x}_1(\omega = 0) - \beta_2 \frac{1}{2}\left|\underline{x}_0(\omega)\right|^2$ or solved for $\underline{x}_1(\omega = 0)$:

$$\underline{x}_1(\omega = 0) = -\frac{1}{2}\frac{\beta_2}{\omega_r^2}\left|\underline{x}_0(\omega)\right|^2 = -\frac{1}{2}\frac{\beta_2}{\omega_r^2}\left(\frac{V\varepsilon_0}{Ne}\right)^2 \left|\underline{E}\right|^2 \left|\underline{\chi}^{(1)}(\omega)\right|^2 \qquad (0.5)$$

- Terms at ω give us:

$$\underline{x}_1(\omega) = -\frac{6}{8}\beta_3 \frac{\left|\underline{x}_0(\omega)\right|^2 \underline{x}_0(\omega)}{\omega_r^2 - \omega + j\omega\gamma_r} = \frac{3}{4}\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^4 \frac{m_e}{e}\underline{E}\left|\underline{E}\right|^2 \left|\underline{\chi}^{(1)}(\omega)\right|^2 \left(\underline{\chi}^{(1)}(\omega)\right)^2 \qquad (0.6)$$

- Terms at 2ω:

$$\underline{x}_1(2\omega) = -\frac{1}{2}\beta_2 \frac{\underline{x}_0(\omega)^2}{\omega_r^2 - (2\omega)^2 + j2\omega\gamma_r} = -\frac{1}{2}\beta_2\left(\frac{V\varepsilon_0}{Ne}\right)^3 \frac{m_e}{e}\underline{E}^2\underline{\chi}^{(1)}(2\omega)\left(\underline{\chi}^{(1)}(\omega)\right)^2 \qquad (0.7)$$

- Terms at 3ω:

$$\underline{x}_1(3\omega) = -\frac{1}{4}\beta_3 \frac{\underline{x}_0(\omega)^3}{\omega_r^2 - (3\omega)^2 + j3\omega\gamma_r} = \frac{1}{4}\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^4 \frac{m}{e}\underline{E}^3\left(\underline{\chi}^{(1)}(\omega)\right)^3 \underline{\chi}^{(1)}(3\omega) \qquad (0.8)$$

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

If we had chosen frequency components that are not contained in the spectrum of the solution, they would have canceled when we separated into terms oscillating at the same frequency.

3.Consider $P(\omega_p) = -\frac{N}{V}e\underline{x}_1(\omega_p)$ and insert the corresponding term for $\underline{x}_1$ from 2.

- Terms at $\omega = 0$ give us:

$$P(\omega = 0) = \frac{N}{V}e\frac{1}{2}\frac{\beta_2}{\omega_r^2}\left|\underline{x}_0(\omega)\right|^2$$

- Terms at ω give us:

$$P(\omega)=\frac{3}{4}\frac{N}{V}e\beta_3\frac{\left|\underline{x}_0(\omega)\right|^2\underline{x}_0(\omega)}{\omega_\mathrm{r}^2-\omega+j\omega\gamma_\mathrm{r}} \tag{0.9}$$

- Terms at 2ω:

$$P(2\omega)=\frac{1}{2}\frac{N}{V}e\beta_2\frac{\underline{x}_0(\omega)^2}{\omega_\mathrm{r}^2-(2\omega)^2+j2\omega\gamma_\mathrm{r}} \tag{0.10}$$

- Terms at 3ω:

$$P(3\omega)=\frac{1}{4}\frac{N}{V}e\beta_3\frac{\underline{x}_0(\omega)^3}{\omega_\mathrm{r}^2-(3\omega)^2+j3\omega\gamma_\mathrm{r}} \tag{0.11}$$

3. We write down the expressions for the nonlinear polarizations:

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

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

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

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

In the very same manner we obtained expressions for the first order susceptibility in the last tutorial by equating the total induced dipole moment and the dielectric polarization, we apply the same scheme to get the respective 2nd and 3rd order susceptibilities:

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

j. $\chi^{(2)}(2\omega:\omega,\omega)=-2\frac{Ne}{V\varepsilon_0}\frac{\underline{x}_1(2\omega)}{\underline{E}^2}=\beta_2\left(\frac{V\varepsilon_0}{Ne}\right)^2\frac{m_\mathrm{e}}{e}\underline{\chi}^{(1)}(2\omega)\left(\underline{\chi}^{(1)}(\omega)\right)^2$

k. $\chi^{(3)}(\omega:\omega,\omega,-\omega)=-\frac{4}{3}\frac{Ne}{V\varepsilon_0}\frac{\underline{x}_1(\omega)}{\underline{E}^2\underline{E}^*}=-\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^3\frac{m_\mathrm{e}}{e}\left|\underline{\chi}^{(1)}(\omega)\right|^2\left(\underline{\chi}^{(1)}(\omega)\right)^2$

l. $\chi^{(3)}(3\omega:\omega,\omega,\omega)=-4\frac{Ne}{V\varepsilon_0}\frac{\underline{x}_1(3\omega)}{\underline{E}^3}=-\beta_3\left(\frac{V\varepsilon_0}{Ne}\right)^3\frac{m}{e}\left(\underline{\chi}^{(1)}(\omega)\right)^3\underline{\chi}^{(1)}(3\omega)$

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