Problem Set 5
Nonlinear Optics (NLO)

Due: May 30, 2018, 08:00 AM

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 and include the damping force $-m_\mathrm{e} \gamma_\mathrm{r} \frac{\mathrm{d}x(t)}{\mathrm{d}t}$:

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

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:

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

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

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, and at zero frequency (DC, i.e. a static field). 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. $\chi^{(3)}(3\omega:\omega,\omega,\omega)$

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