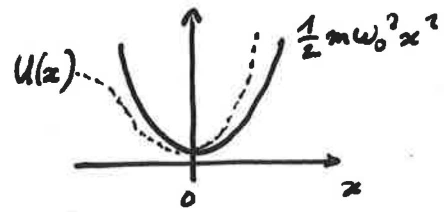
E2 Anharmonic nonlinear oscillator

a. Forces: $\begin{cases} \tilde{F} = -e E_1 e^{-i\omega t} \\ \tilde{F}_s = -k_1 \tilde{x} \underbrace{- k_2 \tilde{x}^2}_{\text{nonlinear restoring force introducing anharmonicity}} \quad (\text{we use order 2 only}) \\ \tilde{F}_d = -2\gamma m \dot{\tilde{x}} \end{cases}$

equation of motion:
$$\ddot{\tilde{x}} + 2\gamma \dot{\tilde{x}} + \underbrace{\frac{k_1}{m}}_{\equiv \omega_0^2} \tilde{x} + \underbrace{\frac{k_2}{m}}_{\equiv a} \tilde{x}^2 = -\frac{e E_1 e^{-i\omega t}}{m} \qquad (1)$$

γ : damping rate
ω_0 : angular frequency of the harmonic oscillator
a : parameter characterizing the strength of the nonlinearity

b. $$U(\tilde{x}) = -\int \tilde{F}_{restoring} \cdot d\tilde{x} = -\int \tilde{F}_s \cdot d\tilde{x}$$
$$= \frac{1}{2} m \omega_0^2 \tilde{x}^2 + \underbrace{\frac{1}{3} m a \tilde{x}^3}_{\text{anharmonicity breaking the energy symmetry}}$$



(case $a > 0$)

c. [Perturbative approach: driving field $\lambda E_1 e^{i\omega t}$ with $\lambda \in [0;1]$
λ characterizes the strength of the perturbation from driving field.

Equation (1) becomes $$\ddot{\tilde{x}} + 2\gamma \dot{\tilde{x}} + \omega_0^2 \tilde{x} + a \tilde{x}^2 = -\frac{\lambda e E_1 e^{-i\omega t}}{m} \qquad (2)$$

We now seek $\tilde{x}$ as a power series expansion in λ:
$$\tilde{x} = \lambda \tilde{x}^{(1)} + \lambda^2 \tilde{x}^{(2)} + \underbrace{(\lambda^3 \tilde{x}^{(3)} + \dots)}_{\text{we neglect this part}}$$

We inject it in (2):
$$\lambda \ddot{\tilde{x}}^{(1)} + \lambda^2 \ddot{\tilde{x}}^{(2)} + 2\gamma [\lambda \dot{\tilde{x}}^{(1)} + \lambda^2 \dot{\tilde{x}}^{(2)}] + \omega_0^2 [\lambda \tilde{x}^{(1)} + \lambda^2 \tilde{x}^{(2)}]$$
$$+ a [\lambda^2 (\tilde{x}^{(1)})^2 + 2\lambda^3 \tilde{x}^{(1)} \cdot \tilde{x}^{(2)} + \lambda^4 (\tilde{x}^{(2)})^2] = -\frac{\lambda e E_1 e^{-i\omega t}}{m}$$

In order for this equation to be a solution for every λ, it has to satisfy equations with terms proportional to λ and λ^2 separately:

here we say $\lambda = 1$ $\begin{cases} \lambda & \ddot{\tilde{x}}^{(1)} + 2\gamma \dot{\tilde{x}}^{(1)} + \omega_0^2 \tilde{x}^{(1)} = -\dfrac{e E_1 e^{-i\omega t}}{m} \quad (3a) \\ \lambda^2 & \ddot{\tilde{x}}^{(2)} + 2\gamma \dot{\tilde{x}}^{(2)} + \omega_0^2 \tilde{x}^{(2)} + a (\tilde{x}^{(1)})^2 = 0 \quad (3b) \end{cases}$

In order to solve this we need to use the following:

ANSATZ $\tilde{x}^{(1)}(t) = x^{(1)}(\omega_1) e^{-i\omega_1 t} + x^{(1)}(\omega_2) e^{-i\omega_2 t} + \dots$

$$= \sum_n x^{(1)}(\omega_n) e^{-i\omega_n t}$$

Note that in our case, the driving field contains only one frequency, thus reducing the number of frequencies in $\tilde{x}(t)$.

(3a) $-\omega_n^2 x^{(1)} e^{-i\omega_n t} - 2\gamma i\omega_n x^{(1)} e^{-i\omega_n t} + \omega_0^2 x^{(1)} e^{-i\omega_n t} = -\frac{eE_1 e^{-i\omega t}}{m}$

Here the driving field produces only a response at frequency ω.

$$\boxed{x^{(1)}(\omega) = \frac{-eE_1}{m(\omega_0^2 - 2\gamma i\omega - \omega^2)} = -\frac{e}{m}\frac{E_1}{D(\omega)}}$$

$$P^{(1)}(\omega) = -Nex^{(1)}(\omega) = \frac{Ne^2 E_1}{mD(\omega)} = \varepsilon_0 \chi^{(1)}(\omega;\omega) E_1(\omega)$$

d. Same reasoning with (3b) equation:

$$-\omega_n^2 x^{(2)} e^{-i\omega_n t} - 2\gamma i\omega_n x^{(2)} e^{-i\omega_n t} + \omega_0^2 x^{(2)} e^{-i\omega_n t} + a\, x^{(1)}(\underline{\underline{\omega_\ell}}) e^{-i\underline{\underline{\omega_\ell}} t} \cdot x^{(1)}(\underline{\underline{\omega_m}}) e^{-i\underline{\underline{\omega_m}} t} = 0$$

Here the driving field imposes $\omega_\ell = \omega_m = \omega$, thus $\omega_n = 2\omega$ to satisfy the equation.

then $\boxed{x^{(2)}(2\omega) = \frac{a\left(-eE_1/mD(\omega)\right)^2}{D(2\omega)} = \frac{ae^2E_1^2}{m^2 D(2\omega) D(\omega)^2}}$ generation of second harmonic

$$P^{(2)}(2\omega) = \varepsilon_0 \chi^{(2)}(2\omega;\omega,\omega) E_1(\omega)^2 = -\frac{Ne^3 a E_1^2}{m^2 D(2\omega) D(\omega)^2}$$

$$\chi^{(2)}(2\omega;\omega,\omega) = -\frac{Ne^3 a}{\varepsilon_0 m^2 D(2\omega) D(\omega)^2}$$

e. Non-resonant condition: $\omega_0 \gg \omega$ (ω is far from ω_0, and ω_0, as a frequency of electronic levels in the material, is big)

Then $\boxed{\chi^{(2)}(2\omega;\omega,\omega) \sim -\frac{Nae^3}{\varepsilon_0 m^2 \omega_0^6}}$

f. We are in 1D so we can use $\chi^{(2)}_{333}$ as approximation for $\chi^{(2)}$.

$$a = -\frac{\chi^{(2)} \varepsilon_0 m^2 \omega_0^6}{Ne^3} [s^{-2} m^{-1}] \Rightarrow \left|\frac{a}{\omega_0^2}\right| \sim 9.5 \times 10^9 \ m^{-1}$$

(strong nonlinear effect)

NB: a and ω_0^2 have different dimensions, thus we cannot say $a \gg \omega_0^2$ but rather $|a/\omega_0^2| \gg 1$