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PROF. ULF PESCHEL

Theory of Nonlinear Optics

All Exercises

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Contents

1 Nonlinear response functions

Consider the nonlinear harmonic oscillator model for the polarization $P(t)$:

$$\ddot{P}(t) + 2\gamma\dot{P}(t) + \omega_0^2 P(t) = \varepsilon_0\omega_f^2 E(t) + \lambda a P^2(t). \tag{1.1}$$

Here $E(t)$ is the external driving field and ω_f is the oscillation strength. The parameter a characterizes the strength of the nonlinear response and $\lambda \ll 1$ is a perturbation parameter. Also for simplicity we have neglected all polarization effects

Exercise 1: *Expand $P(t)$ in powers of λ, i. e. $P(t) = P^{(1)}(t) + \lambda P^{(2)}(t) + \mathcal{O}(\lambda^2)$, insert that expression into above equation and find the equations which determine $P^{(1)}(t)$ and $P^{(2)}(t)$ by putting the terms in front of the different powers of λ to zero.*

Solution: Substituting the Ansatz into equation (1.1) and neglecting all orders $\mathcal{O}(\lambda^2)$ we find

$$\ddot{P}^{(1)} + \lambda\ddot{P}^{(2)} + 2\gamma\dot{P}^{(1)} + 2\gamma\lambda\dot{P}^{(2)} + \omega_0^2 P^{(1)} + \omega_0^2\lambda P^{(2)} = \varepsilon_0\omega_f^2 E(t) + \lambda a {P^{(1)}}^2. \tag{1.2}$$

By comparing the different orders of λ we find

$$\ddot{P}^{(1)} + 2\gamma\dot{P}^{(1)} + \omega_0^2 P^{(1)} = \varepsilon_0\omega_f^2 E(t) \tag{1.3}$$

$$\ddot{P}^{(2)} + 2\gamma\dot{P}^{(2)} + \omega_0^2 P^{(2)} = a {P^{(1)}}^2. \tag{1.4}$$

Exercise 2: *Find the first order response function $R^{(1)}(t)$ defined as*

$$\ddot{R}^{(1)}(t) + 2\gamma\dot{R}^{(1)}(t) + \omega_0^2 R^{(1)}(t) = \omega_f^2\delta(t), \qquad R^{(1)}(t) = 0 \quad \textit{for} \quad t < 0, \tag{1.5}$$

and show that the first order polarization can be expressed as

$$P^{(1)}(t) = \varepsilon_0 \int_{-\infty}^{\infty} R^{(1)}(\tau_1) E(t-\tau_1)\,\mathrm{d}\tau_1\,. \tag{1.6}$$

Solution: We can solve this second order, inhomogenous, linear differential equation by first solving the homogenous part. We do that by making the ansatz $R^{(1)}(t) = C\exp(\lambda t)$ and substituting it into (1.5)

$$\lambda^2 + 2\gamma\lambda + \omega_0^2 = 0 \quad \Rightarrow \quad \lambda_{1/2} = -\gamma \pm \sqrt{\gamma^2 - \omega_0^2}. \tag{1.7}$$

With two λ's the general solution is

$$R^{(1)}(t) = C_1 \mathrm{e}^{\lambda_1 t} + C_2 \mathrm{e}^{\lambda_2 t}. \tag{1.8}$$

We can find the particula solution by varying the constants C_1 and C_2. For the problem

$$R_p^{(1)}(t) = C_1(t)R_1(t) + C_2(t)R_2(t) \quad \text{with} \quad a\ddot{R}(t) + b\dot{R}(t) + c\dot{R}(t) = F(t) \tag{1.9}$$

the generic solution for the constants is[1]

$$\dot{C}_1(t) = -\frac{1}{a}F(x)\frac{R_2}{R_1\dot{R}_2 - R_2\dot{R}_1}, \qquad \dot{C}_2(t) = +\frac{1}{a}F(x)\frac{R_1}{R_1\dot{R}_2 - R_2\dot{R}_1}. \tag{1.10}$$

Using this we find

$$\begin{aligned} \dot{C}_1(t) &= -\omega_f^2\delta(t)\frac{e^{\lambda_2 t}}{e^{(\lambda_1+\lambda_2)t}(\lambda_2-\lambda_1)} = -\frac{\omega_f^2\delta(t)}{\lambda_2-\lambda_1}e^{-\lambda_1 t} \\ \dot{C}_2(t) &= +\omega_f^2\delta(t)\frac{e^{\lambda_1 t}}{e^{(\lambda_1+\lambda_2)t}(\lambda_2-\lambda_1)} = +\frac{\omega_f^2\delta(t)}{\lambda_2-\lambda_1}e^{-\lambda_2 t}. \end{aligned} \tag{1.11}$$

Integrating the two equations requires performing the following integral

$$\int_{-\infty}^{\infty} \delta(t)e^{-\lambda_i t}\,dt = e^{-\lambda_i 0} = 1. \tag{1.12}$$

Therefore we have

$$C_1(t) = -\frac{\omega_f^2}{\lambda_2-\lambda_1} = +\frac{1}{2}\frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}} = -C_2(t). \tag{1.13}$$

Therefore the general solution can be written as

$$\begin{aligned} R^{(1)}(t) &= \frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}}\frac{1}{2}\left[e^{\lambda_1 t} - e^{\lambda_2 t}\right] = \frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}}\frac{1}{2}e^{-\gamma t}\left[e^{\sqrt{\gamma^2-\omega_0^2}\,t} - e^{-\sqrt{\gamma^2-\omega_0^2}\,t}\right] \\ &= \frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}}e^{-\gamma t}\sinh\left(\sqrt{\gamma^2-\omega_0^2}\,t\right). \end{aligned} \tag{1.14}$$

We can now solve (1.3) in the same way by replacing $\omega_f^2\delta(t) \to \varepsilon_0\omega_f^2 E(t)$

$$P^{(1)}(t) = C_1(t)e^{\lambda_1 t} + C_2 e^{\lambda_2 t}, \qquad \dot{C}_{1/2} = \pm\frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}}\varepsilon_0 E(t)e^{-\lambda_{1/2}t}. \tag{1.15}$$

[1] c.f. Lotze, Mathematische Methoden der Physik 1 (script), p. 33

The general solution can now be written as

$$\begin{aligned}
P^{(1)}(t) &= \varepsilon_0 \int_{-\infty}^{\infty} \frac{1}{2}\frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}} E(\tau)\left[e^{\lambda_1(t-\tau)} - \mathrm{e}^{\lambda_2(t-\tau)}\right]\mathrm{d}\tau \\
&= \varepsilon_0 \int_{-\infty}^{\infty} \underbrace{\frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}} \mathrm{e}^{\gamma(t-\tau)} \sinh\left(\sqrt{\gamma^2-\omega_0^2}(t-\tau)\right)}_{R^{(1)}(t-\tau)} E(\tau)\,\mathrm{d}\tau \qquad \tau_1 = t-\tau \\
&= \varepsilon_0 \int_{-\infty}^{\infty} R^{(1)}(\tau_1) E(t-\tau_1)\,\mathrm{d}\tau_1 .
\end{aligned} \tag{1.16}$$

Exercise 3 *What is* $\chi^{(1)}(\omega)$*?*

Solution: The susceptibility $\chi^{(1)}(\omega)$ is defined as the Fourier transform of the response function $R^{(1)}(t)$

$$\begin{aligned}
\chi^{(1)}(\omega) &= \int_{-\infty}^{\infty} R^{(1)}(t)\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}t = \int_{0}^{\infty} \frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}} \mathrm{e}^{-\gamma t}\sinh\left(\sqrt{\gamma^2-\omega_0^2}\,t\right)\mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}t \\
&= \frac{\omega_f^2}{\sqrt{\gamma^2-\omega_0^2}} \int_0^{\infty} \frac{1}{2}\mathrm{e}^{(\mathrm{i}\omega-\gamma)t}\left[\mathrm{e}^{\sqrt{\gamma^2-\omega_0^2}\,t} - \mathrm{e}^{-\sqrt{\gamma^2-\omega_0^2}\,t}\right]\mathrm{d}t \\
&= \frac{\omega_f^2}{2\sqrt{\gamma^2-\omega_0^2}}\left[\frac{\mathrm{e}^{\sqrt{\gamma^2-\omega_0^2}\,t}}{\mathrm{i}\omega-\gamma+\sqrt{\gamma^2-\omega_0^2}} - \frac{\mathrm{e}^{-\sqrt{\gamma^2-\omega_0^2}\,t}}{\mathrm{i}\omega-\gamma-\sqrt{\gamma^2-\omega_0^2}}\right]\mathrm{e}^{(\mathrm{i}\omega-\gamma)t}\Big|_0^{\infty} \\
&= -\frac{\omega_f^2}{2\sqrt{\gamma^2-\omega_0^2}}\left[\frac{1}{\mathrm{i}\omega-\gamma+\sqrt{\gamma^2-\omega_0^2}} - \frac{1}{\mathrm{i}\omega-\gamma-\sqrt{\gamma^2-\omega_0^2}}\right] \\
&= -\frac{\omega_f^2}{2\sqrt{\gamma^2-\omega_0^2}}\left[\frac{\mathrm{i}\omega-\gamma-\sqrt{\gamma^2-\omega_0^2}}{(\mathrm{i}\omega-\gamma)^2-(\gamma^2-\omega_0^2)} - \frac{\mathrm{i}\omega-\gamma+\sqrt{\gamma^2-\omega_0^2}}{(\mathrm{i}\omega-\gamma)^2-(\gamma^2-\omega_0^2)}\right] \\
&= -\frac{\omega_f^2}{2\sqrt{\gamma^2-\omega_0^2}}\left[\frac{-2\sqrt{\gamma^2-\omega_0^2}}{(\mathrm{i}\omega-\gamma)^2-\gamma^2+\omega_0^2}\right] = \frac{\omega_f^2}{\omega_0^2-\omega^2-\mathrm{i}2\gamma\omega}.
\end{aligned} \tag{1.17}$$

We can further split the result into its real and imaginary part

$$\chi^{(1)}(\omega) = \frac{\omega_f^2(\omega_0^2-\omega^2)}{(\omega_0^2-\omega^2)^2+4\gamma^2\omega^2} + \mathrm{i}\frac{\omega_f^2 2\gamma\omega}{(\omega_0^2-\omega^2)^2+4\gamma^2\omega^2}. \tag{1.18}$$

Both parts are depicted in figure 1.

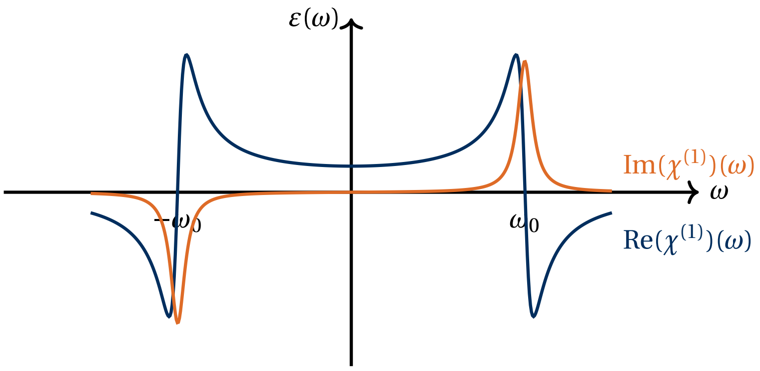

Fig. 1: Real and imaginary part of the linear susceptibility $\chi^{(1)}(\omega)$.

Exercise 4 *Express P^2 in terms of $P^{(1)}$ using the response function $R^{(1)}$ defined above. Then replace $P^{(1)}$ using expression (1.6) and construct an expression giving $P^{(2)}$ as a function of the electric field.*

Solution: Analagous to exercise two we can solve the differential equation (1.4) easily by substituting $\omega_f^2\delta(t) \to aP^{(1)^2}$ which leads to

$$P^{(2)}(t) = C_1(t)\mathrm{e}^{\lambda_1 t} + C_2\mathrm{e}^{\lambda_2 t}, \qquad \dot{C}_{1/2} = \pm\frac{aP^{(1)^2}(t)}{\sqrt{\gamma^2-\omega_0^2}}\mathrm{e}^{-\lambda_{1/2}t}. \tag{1.19}$$

The general solution can now be written as

$$\begin{aligned}
P^{(2)}(t) &= \int_{-\infty}^{\infty} \frac{1}{2}\frac{a}{\sqrt{\gamma^2-\omega_0^2}}P^{(1)^2}(\tau)\left[e^{\lambda_1(t-\tau)} - \mathrm{e}^{\lambda_2(t-\tau)}\right]\mathrm{d}\tau \\
&= \int_{-\infty}^{\infty} \frac{a}{\sqrt{\gamma^2-\omega_0^2}}P^{(1)^2}(\tau)\mathrm{e}^{\gamma(t-\tau)}\sinh\left(\sqrt{\gamma^2-\omega_0^2}(t-\tau)\right)\mathrm{d}\tau \\
&= \int_{-\infty}^{\infty} \frac{a}{\omega_f^2}\underbrace{P^{(1)^2}(t-\tau)}R^{(1)}(\tau)\,\mathrm{d}\tau \quad (indexshift) \\
&\qquad\qquad = \left(\varepsilon_0\int_{-\infty}^{\infty} R^{(1)}(\tau_1')E(t-\tau-\tau_1')\,\mathrm{d}\tau_1'\right)^2 \\
&= \varepsilon_0\int_{-\infty}^{\infty}\frac{\varepsilon_0 a}{\omega_f^2}\left(\iint_{-\infty}^{\infty}\mathrm{d}\tau_1'\,\mathrm{d}\tau_2'\,R^{(1)}(\tau_1')R^{(1)}(\tau_2')E(t-\tau-\tau_1')E(t-\tau-\tau_2')\right)\mathrm{d}\tau \quad \tau_i = \tau_i' + \tau \\
&= \varepsilon_0\iiint_{-\infty}^{\infty}\frac{\varepsilon_0 a}{\omega_f^2}R^{(1)}(\tau_1-\tau)R^{(1)}(\tau_2-\tau)R^{(1)}(\tau)E(t-\tau_1)E(t-\tau_2)\,\mathrm{d}\tau\,\mathrm{d}\tau_1\,\mathrm{d}\tau_2\,.
\end{aligned} \tag{1.20}$$

Exercise 5 *Express the second order response function $R^{(2)}(\tau_1, \tau_2)$, defined as*

$$P^{(2)}(t) = \varepsilon_0 \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} R^{(2)}(\tau_1, \tau_2) E(t-\tau_1) E(t-\tau_2)\, \mathrm{d}\tau_1\, \mathrm{d}\tau_2 \tag{1.21}$$

in terms of $R^{(1)}(t)$.

Solution: Comparing both equations for the second order polarization (1.20) and (1.21) we find

$$R^{(2)}(\tau_1, \tau_2) = \int_{-\infty}^{\infty} \frac{\varepsilon_0 a}{\omega_f^2} R^{(1)}(\tau) R^{(1)}(\tau_1 - \tau) R^{(1)}(\tau_2 - \tau)\, \mathrm{d}\tau\,. \tag{1.22}$$

2 Third order nonlinear response

Exercise 1: *Consider the third order nonlinear polarization $\boldsymbol{P}^{(3)}$ in an isotropic medium with Kleinmann symmetry. Show*

a) that the ellipticity of $\boldsymbol{P}^{(3)}$ may differ from that of $\boldsymbol{E}$

b) that in some cases $\boldsymbol{P}^{(3)}$ is not parallel to $\boldsymbol{E}$

c) that the induced effective nonlinear susceptibility is different for circularly and linearly polarized waves, and

d) that losses occur, if $\mathrm{Im}(\chi^{(3)}) > 0$ holds.

Solution:

Exercise 2: *Surface second harmonic generation is caused by a quadratic nonlinearity and occurs even at the suraces of materials as e. g. silicon, which possess no quadratic nonlinearity and occurs even at the surfaces of materials as e. g. silicon, which possesses no quadratic bulk nonlinearity. We now investigate second harmonic generation at the flat surface of an isotropic material lying in the x-y-plane.*

a) Why is the occurrence of surface second harmonic in isotropic materials no contradiction to the absence of a respective bulk response?

b) Which tensor elements of $\chi^{(2)}_{ijk}(-2\omega|\omega,\omega)$ are nonzero at the interface? How many independent coefficients exist?

Exercise 3: *Assume two linearly polarized plane optical waves*

$$\boldsymbol{E}_{1/2}(\boldsymbol{r},t) = \frac{1}{2}\left\{E_{1/2}\hat{\mathbf{e}}_x \exp[\mathrm{i}(k_{1/2}z - \omega_{1/2}t)] + c.c.\right\} \tag{2.1}$$

to propagate in an isotropic material at two distinct frequencies. Derive the terms of the cubic nonlinear polarization $\boldsymbol{P}^{(3)}$, which oscillate at frequencies ω_1 and ω_2. To simplify the derivation make use of Kleinmann symmetry. How do the two waves influence each other, if the generation of new frequencies can be neglected?

Solution: Using the general expression of the third order polarization we can write

$$\boldsymbol{P}^{(3)}(t) = \frac{1}{2}\sum_{\omega_p}\left(\boldsymbol{P}^{(3)}_{\omega_p}\mathrm{e}^{k_p z - \mathrm{i}\omega_p t} + \mathrm{c.c.}\right), \tag{2.2}$$

where ω_p are all possible combinations of the frequencies ω_1 and ω which are

$$\omega_p = \{\omega_1, \omega_2, 3\omega_1, 3\omega_2, 2\omega_1 + \omega_2, \omega_1 + 2\omega_2, 2\omega_1 - \omega_2, 2\omega_2, -\omega_1\}. \tag{2.3}$$

We now neglect all new frequencies $\omega_p \neq \omega_1, \omega_2$. Furthermore we also assume Kleinmann symmetry which means

$$\chi^{(3)}(\omega_1) = \chi^{(3)}(\omega_2). \tag{2.4}$$

We can now rewrite equation (2.2) as

$$\begin{aligned}
\boldsymbol{P}^{(3)}(t) &= \frac{1}{2}\chi^{(3)}\varepsilon_0 \sum_{\omega_p} \Big(K(-\omega_p|\omega_\alpha,\omega_\beta,\omega_\gamma) \mathrm{e}^{k_p z - \mathrm{i}\omega_p t} E^{(*)}_{\alpha,\omega_p} E^{(*)}_{\beta,\omega_p} E^{(*)}_{\gamma,\omega_p} + \text{c.c.} \Big) \\
&= \frac{1}{2}\chi^{(3)}\varepsilon_0 \big[K(\omega_1|\omega_1,\omega_1,-\omega_1)\mathrm{e}^{k_1 z - \mathrm{i}\omega_1 t} E_1^2 E_1^* \\
&\qquad K(\omega_1|\omega_1,\omega_2,-\omega_2)\mathrm{e}^{k_1 z - \mathrm{i}\omega_1 t} E_1 E_2 E_2^* \\
&\qquad K(\omega_2|\omega_2,\omega_1,-\omega_1)\mathrm{e}^{k_2 z - \mathrm{i}\omega_2 t} E_1 E_2 E_1^* \\
&\qquad K(\omega_1|\omega_2,\omega_2,-\omega_2)\mathrm{e}^{k_2 z - \mathrm{i}\omega_2 t} E_2 E_2 E_2^* + \text{c.c.} \big].
\end{aligned}$$

Now we simply have to evaluate the $K(\ldots)$ factors by using

$$K(-\omega_p|\omega_1,\ldots,\omega_n) = 2^{l+m-n} p = \frac{p}{4} \tag{2.5}$$

where $n = 3$ is the order of the interaction, p the number of permutations, m the number of ω_i being zero and $l = 1$ because $\omega_p \neq 0$. For $K(\omega_1|\omega_1,\omega_1,-\omega_1)$ we can find three possible permutations for $-\omega_1$ being in the three different positions. For $K(\omega_1|\omega_1,\omega_2,-\omega_2)$ all frequencies are different which means $p = n! = 6$.

We can now conclude:

$$\boldsymbol{P}^{(3)}(t) = \frac{1}{8}\chi^{(3)}\varepsilon_0 \Big[(3E_1^2 E_1^* + 6E_1 E_2 E_2^*)\mathrm{e}^{k_1 z - \mathrm{i}\omega_1 t} + (3E_2^2 E_2^* + 6E_2 E_1 E_1^*)\mathrm{e}^{k_2 z - \mathrm{i}\omega_2 t} \Big]. \tag{2.6}$$

We can verify this result by using the *experimentalist approach* of calculating the polarization, where we define the total electric field (neglecting the spatial dependence) as

$$\boldsymbol{E}(t) = \frac{1}{2}\big[E_1 \mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2 \mathrm{e}^{-\mathrm{i}\omega_2 t} + \text{c.c.} \big] \tag{2.7}$$

and calculate $P^{(3)}(t)$ in the following way:

$$\begin{aligned}
P^{(3)}(t) &= \frac{1}{8}\varepsilon_0\chi^{(3)} \big[E_1 \mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2 \mathrm{e}^{-\mathrm{i}(\omega_2) t} + c.c. \big]^3 \\
&= \frac{1}{8}\varepsilon_0\chi^{(3)} \big[E_1 \mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2 \mathrm{e}^{-\mathrm{i}\omega_2 t} + E_1^* \mathrm{e}^{\mathrm{i}\omega_1 t} + E_2^* \mathrm{e}^{\mathrm{i}(\omega_2) t} \big]^3 \\
&= \frac{1}{8}\varepsilon_0\chi^{(3)} \big[(E_1 \mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2 \mathrm{e}^{-\mathrm{i}\omega_2 t})^3 + 3(E_1 \mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2 \mathrm{e}^{-\mathrm{i}\omega_2 t}) \cdot (E_1^* \mathrm{e}^{\mathrm{i}\omega_1 t} + E_2^* \mathrm{e}^{\mathrm{i}(\omega_2) t}) + c.c. \big] \\
&= \frac{1}{8}\varepsilon_0\chi^{(3)} \big[E_1^2 \mathrm{e}^{-\mathrm{i}3\omega_1 t} + 3E_1^2 E_2 \mathrm{e}^{-\mathrm{i}(2\omega_1+\omega_2) t} + 3E_1 E_2^2 \mathrm{e}^{-\mathrm{i}(2\omega_2+\omega_1) t} + 3E_2^3 \mathrm{e}^{-\mathrm{i}3\omega_2 t} \\
&\qquad + 3E_1^2 E_1^* \mathrm{e}^{-\mathrm{i}\omega_1 t} + 6E_1 E_2 E_1^* \mathrm{e}^{-\mathrm{i}\omega_2 t} + 3E_2^2 E_1^* \mathrm{e}^{-\mathrm{i}(2\omega_2-\omega_1) t} \\
&\qquad + 3E_1^2 E_2^* \mathrm{e}^{-\mathrm{i}(2\omega_1-\omega_2) t} + 6E_1 E_2 E_2^* \mathrm{e}^{-\mathrm{i}\omega_1 t} + 3E_2^2 E_2^* \mathrm{e}^{-\mathrm{i}\omega_2 t} + c.c. \big]
\end{aligned} \tag{2.8}$$

3 Two level systems

Exercise 1: *Show that the matrix element of the momentum operator $\hat{\boldsymbol{p}}$ with respect to energy eigenstates $\langle\psi_a|\hat{\boldsymbol{p}}|\psi_b\rangle$ can be expressed in terms of the position operator $\hat{\boldsymbol{r}}$ as*

$$\langle\psi_a|\hat{\boldsymbol{p}}|\psi_b\rangle = \mathrm{i}\frac{m}{\hbar}(E_a - E_b)\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle \tag{3.1}$$

where $|\psi_{a/b}\rangle$ are eigenstates of the Hamiltonian $\hat{H}_0 = \frac{\hat{\boldsymbol{p}}^2}{2m} + V(\hat{\boldsymbol{r}})$ with $\hat{H}_0|\psi_{a/b}\rangle = E_{a/b}|\psi_{a/b}\rangle$. Hint: Make use of the commutator $[\hat{\boldsymbol{r}}, \hat{H}_0]$.

Solution: First we try to calculate the commutator $[\hat{\boldsymbol{r}}, \hat{H}_0]$:

$$\begin{aligned}
[\hat{\boldsymbol{r}}, \hat{H}_0] &= \hat{\boldsymbol{r}}\left(\frac{\hat{\boldsymbol{p}}^2}{2m} + \cancel{V(\hat{\boldsymbol{r}})}\right) - \left(\frac{\hat{\boldsymbol{p}}^2}{2m} + \cancel{V(\hat{\boldsymbol{r}})}\right)\hat{\boldsymbol{r}} \\
&= \frac{1}{2m}\left(\hat{\boldsymbol{r}}\hat{\boldsymbol{p}}^2 - \hat{\boldsymbol{p}}^2\hat{\boldsymbol{r}}\right) \\
&= \frac{1}{2m}\Big(\underbrace{\hat{\boldsymbol{r}}\hat{\boldsymbol{p}}\hat{\boldsymbol{p}} - \hat{\boldsymbol{p}}\hat{\boldsymbol{r}}\hat{\boldsymbol{p}}}_{=[\hat{\boldsymbol{r}},\hat{\boldsymbol{p}}]\hat{\boldsymbol{p}}} + \underbrace{\hat{\boldsymbol{p}}\hat{\boldsymbol{r}}\hat{\boldsymbol{p}} - \hat{\boldsymbol{p}}\hat{\boldsymbol{p}}\hat{\boldsymbol{r}}}_{\hat{\boldsymbol{p}}[\hat{\boldsymbol{r}},\hat{\boldsymbol{p}}]}\Big) \\
&= \frac{1}{2m}\Big(\underbrace{[\hat{\boldsymbol{r}}, \hat{\boldsymbol{p}}]}_{=\mathrm{i}\hbar}\hat{\boldsymbol{p}} + \hat{\boldsymbol{p}}\underbrace{[\hat{\boldsymbol{r}}, \hat{\boldsymbol{p}}]}_{=\mathrm{i}\hbar}\Big) \\
&= \frac{\mathrm{i}\hbar}{2m}(\hat{\boldsymbol{p}} + \hat{\boldsymbol{p}}) = \frac{\mathrm{i}\hbar}{m}\hat{\boldsymbol{p}}.
\end{aligned} \tag{3.2}$$

Therefore we can rewrite the momentum operator $\hat{\boldsymbol{p}}$ as

$$\hat{\boldsymbol{p}} = \frac{m}{\mathrm{i}\hbar}[\hat{\boldsymbol{r}}, \hat{H}_0] \tag{3.3}$$

which can be inserted into the matrix element and use the hermiticity of the Hamiltonian $H_0 = H_0^\dagger \Rightarrow \langle\psi_a|\hat{H}_0^\dagger = \langle\psi_a|\hat{H}_0 = \langle\psi_a|E_a$

$$\begin{aligned}
\langle\psi_a|\hat{\boldsymbol{p}}|\psi_b\rangle &= \frac{m}{\mathrm{i}\hbar}\langle\psi_a|[\hat{\boldsymbol{r}}, \hat{H}_0]|\psi_b\rangle \\
&= \frac{m}{\mathrm{i}\hbar}\left(\langle\psi_a|\hat{\boldsymbol{r}}\hat{H}_0|\psi_b\rangle - \langle\psi_a|\hat{H}_0\hat{\boldsymbol{r}}|\psi_b\rangle\right) \\
&= \frac{m}{\mathrm{i}\hbar}\left(E_b\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle - E_a\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle\right) \\
&= \mathrm{i}\frac{m}{\hbar}(E_a - E_b)\langle\psi_a|\hat{\boldsymbol{r}}|\psi_b\rangle.
\end{aligned} \tag{3.4}$$

Exercise 2: *A two level system is excited by an external optical field. Show that for the lossless case described by the evolution equations*

$$\mathrm{i}\frac{\mathrm{d}\boldsymbol{P}}{\mathrm{d}t} = \Delta\omega\boldsymbol{P} + \frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}I \tag{3.5}$$

$$\frac{\mathrm{d}I}{\mathrm{d}t} = \frac{1}{2\mathrm{i}\hbar N}\boldsymbol{E}^*\boldsymbol{P} + c.c. \tag{3.6}$$

a conservation law like

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\gamma|\boldsymbol{P}|^2 + I^2\right) = 0 \tag{3.7}$$

is valid and determine γ!

Solution: We use the derived evolution equations for the Polarization $\boldsymbol{P}$ and the inversion $I = |a(t)|^2 - |b(t)|^2$ with $|\psi\rangle = a(t)|\psi_a\rangle + b(t)|\psi_b\rangle$ which were derived in the lecture. We start by evaluating the left hand side of equation (3.7) using $|\boldsymbol{P}|^2 = \boldsymbol{P}\boldsymbol{P}^*$. For that we first need the derivative of $\boldsymbol{P}^*$ which can be obtained by using equation (3.5)

$$\frac{\mathrm{d}\boldsymbol{P}^*}{\mathrm{d}t} = -\frac{1}{\mathrm{i}}\left(\Delta\omega\boldsymbol{P}^* + \frac{Ne^2}{\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}^*I\right). \tag{3.8}$$

We note that the inversion I is per definition a real valued quantity. Now we write the left hand side of equation (3.7) as

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\left(\gamma|\boldsymbol{P}|^2 + I^2\right) &= \gamma\boldsymbol{P}^*\frac{\mathrm{d}\boldsymbol{P}}{\mathrm{d}t} + \gamma\boldsymbol{P}\frac{\mathrm{d}\boldsymbol{P}^*}{\mathrm{d}t} + 2I\frac{\mathrm{d}I}{\mathrm{d}t}\\
&= +\cancel{\frac{\gamma\Delta\omega}{i}|\boldsymbol{P}|^2} + \frac{\gamma Ne^2}{\mathrm{i}\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}I\boldsymbol{P}^*\\
&\quad -\cancel{\frac{\gamma\Delta\omega}{i}|\boldsymbol{P}|^2} - \frac{\gamma Ne^2}{\mathrm{i}\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2\boldsymbol{E}^*I\boldsymbol{P}\\
&\quad + \frac{I}{\mathrm{i}\hbar N}\boldsymbol{E}^*\boldsymbol{P} - \frac{I}{\mathrm{i}\hbar N}\boldsymbol{E}\boldsymbol{P}^*\\
&= \frac{\gamma Ne^2}{\mathrm{i}\hbar}|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2 I(\boldsymbol{E}\boldsymbol{P}^* - \boldsymbol{E}^*\boldsymbol{P}) - \frac{I}{\mathrm{i}\hbar N}(\boldsymbol{E}\boldsymbol{P}^* - \boldsymbol{E}^*\boldsymbol{P})\\
&= \underbrace{(\gamma N^2e^2|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2 - 1)}_{=0}\frac{I}{\mathrm{i}\hbar N}(\boldsymbol{E}\boldsymbol{P}^* - \boldsymbol{E}^*\boldsymbol{P}) \overset{!}{=} 0.\end{aligned} \tag{3.9}$$

From that condition we can deduce γ as

$$\gamma = \frac{1}{N^2e|\langle\psi_b|\hat{\boldsymbol{r}}|\psi_a\rangle|^2}. \tag{3.10}$$