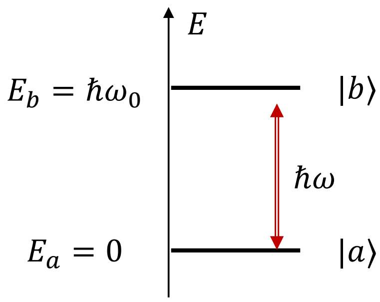
Problem set 04

Nonlinear Optics for Quantum Technologies

March 13, 2025

Nonlinear response of a two-level system (TLS)

We consider a two level system, e.g an atom, as schematically depicted below:



We recall the notation for the density operator $\hat{\rho}$, in the basis of eigenstates $|a\rangle$, $|b\rangle$:

$$\hat{\rho} = \begin{pmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ab}^* & \rho_{bb} \end{pmatrix} \tag{1}$$

Note in particular that $\rho_{ba} = \rho_{ab}^*$

1. Write the time-independent term $\hat{H}_0$ and perturbation term $\hat{H}_1(t)$ of the Hamiltonian when an external field $\vec{E}(t) = E_0 \cos(\omega t)\vec{e}_x$ interacts with the dipole moment operator $\hat{\mu}_x$ of the atom.

   Hints:
   - Introduce the Rabi frequency Ω_1, defined as $\Omega_1 = -\mu_{ab}E_0/\hbar$.
   - Write the explicit definition of μ_{ab}.

2. We recall that the unitary evolution of the system is described by $i\hbar\dot{\hat{\rho}} = \left[\hat{H}, \hat{\rho}\right]$. Write the 4 corresponding equations for the matrix elements $\rho_{ij}(t)$.

3. What is the value of $\frac{d}{dt}(\rho_{aa} + \rho_{bb})$? Interpret in terms of $Tr\left[\hat{\rho}\right]$. How many of the equations derived in question 2 are independent?

4. In the limit $\Omega_1 = 0$, what is the time evolution of each component $\rho_{ij}(t)$?

5. Make the change of variable $\rho'_{ab} = \rho_{ab}e^{-i\omega t}$ and rewrite the equations obtained in question 2. Expand $\cos(\omega t)$ in complex form and retain only slow rotating terms when $\omega \approx \omega_0$ to simplify the equations. This approximation is known as the **rotating-wave approximation**. When is it valid? Justify the change of variable used above.

6. For a closed TLS, relaxation is modeled by additional terms in the time evolution of the density matrix:

$$\{\dot{\rho}_{bb}\}_{rel} = -\Gamma_{\text{sp}}\,\rho_{bb} \tag{2}$$
$$\{\dot{\rho}_{ab}\}_{rel} = -\gamma\,\rho_{ab} \tag{3}$$

   with $\Gamma_{\text{sp}}, \gamma \in \mathbb{R}$. What is the physical meaning of $\{\dot{\rho}_{bb}\}_{rel}$ and $\{\dot{\rho}_{ab}\}_{rel}$? What about $\{\dot{\rho}_{aa}\}_{rel}$ and $\{\dot{\rho}_{ba}\}_{rel}$?

7. Include relaxation terms from question 6 into the equations for ρ_{aa}, ρ_{bb} and ρ'_{ab}.

8. Find the steady-state solutions for ρ_{bb} and ρ'_{ab}.

   Hint: the term $\Omega_1^2\gamma/\Gamma_{\rm sp}$ should appear in the result.

9. Going back from ρ'_{ab} to ρ_{ab}, express the expectation value of the dipole moment $\langle\hat{\mu}_x\rangle = Tr\left[\hat{\rho}\hat{\mu}_x\right]$. Check that you obtain a real number.

10. For a dilute gas with N/V atoms per unit volume, express the real χ' and imaginary χ'' parts of the susceptibility.

11. How do the expressions above depend on the external field ? What happens to absorption upon increasing power?

    Reminder: $n(\omega) = \sqrt{1+\chi(\omega)} \approx 1 + \frac{1}{2}\chi(\omega)$

12. The rates γ and $\Gamma_{\rm sp}$ are actually related as follows:

$$\gamma = \Gamma_{\rm sp}/2 + \gamma_{\rm pd} \tag{4}$$

    where $\gamma_{\rm pd}$ is called the pure dephasing rate.

    How does the value of $\gamma/\Gamma_{\rm sp}$ impact the nonlinear response? For what value of $\gamma_{\rm pd}$ is the nonlinear response easiest to observe? What processes can cause $\gamma_{\rm pd} \neq 0$?