1 Radiation Reaction

The field acting back on the dipole due to radiation is called $\vec{E}_{\text{self}}$ and it can be seen as a **friction**.

$$\vec{F}_R = q\vec{E}_{\text{self}}. \tag{1}$$

The **Abraham Lorentz back action** can be written as

$$\vec{F}_R = \frac{q^2 \dddot{\vec{r}}}{6\pi\epsilon_0 c^3}, \tag{2}$$

where $\vec{\mu} = q\vec{r}$ is the electric dipole moment,

$$\vec{\mu} = q\vec{r} \implies \vec{\mu} = \vec{\mu}_0 e^{-i\omega t},$$

hence

$$\dddot{\vec{r}} = \frac{\vec{\mu}_0}{q}(-i\omega)(-i\omega)(-i\omega)e^{-i\omega t},$$

$$q\dddot{\vec{r}} = i\omega^3\vec{\mu}.$$

Thus we can write $\vec{E}_{\text{self}}$ as follows

$$\vec{E}_{\text{self}} = \frac{i\omega^3\vec{\mu}}{6\pi\epsilon_0 c^3} = \frac{ik^3}{6\pi\epsilon_0}\vec{\mu}.$$

The dipole moment is induced by the polarizability times the applied electric field. The latter is the sum of the external electric field $\vec{E}_0$ and of the back action due to radiation $\vec{E}_{\text{self}}$, i.e.

$$\vec{\mu} = \alpha\Big[\vec{E}_0 + \vec{E}_{\text{self}}\Big].$$

Using the expression for $\vec{E}_{\text{self}}$ we can write

$$\vec{\mu} = \alpha\Big[\vec{E}_0 + \frac{ik^3}{6\pi\epsilon_0}\vec{\mu}\Big],$$

hence

$$\vec{\mu} = \frac{\alpha}{1 - \Big[\frac{ik^3}{6\pi\epsilon_0}\alpha\Big]}\vec{E}_0,$$

which leads to an expression for the effective polarizability with radiative corrections

$$\alpha_{\text{eff}} = \frac{\alpha}{1 - \Big[\frac{ik^3}{6\pi\epsilon_0}\alpha\Big]}.$$

2 Polarizability of a Classical Point Like Radiator

Consider a dipole of charge q and dipole moment $\vec{\mu}$. The equation of motion under an applied electric field $\vec{E} = \vec{E}_0 e^{-i\omega t}$ reads

$$\ddot{\vec{r}} + \Gamma' \dot{\vec{r}} + \tau \dddot{\vec{r}} + \omega_0^2 \vec{r} = \frac{e}{m} \vec{E}_0 e^{-i\omega t},$$

where Γ' represents non-radiative damping, ω_0 is the resonance frequency and $\Gamma = \tau\omega_0^2$ is the radiation damping. By comparing this expression with the Abraham Lorentz back action (see Eq. (2)), we get the expression for the **Radiative Decay Rate** of a classical oscillating dipole

$$\Gamma = \frac{2e^2\omega_0^2}{3mc^3}.$$

The **Steady State Solution** can be written as follows

$$\vec{\mu} = \vec{\mu}_0(\omega) e^{-i\omega t - \Gamma_{\text{tot}}},$$

where $\Gamma_{\text{tot}} = \Gamma' + \frac{\omega^2}{\omega_0^2}\Gamma$ is the total decay rate. Knowing that $\vec{\mu} = -e\vec{r}$, we get

$$\vec{\mu}_0(\omega) = -\frac{e^2}{m} \frac{\vec{E}_0}{\omega_0^2 - \omega^2 - i\omega\Gamma_{\text{tot}}}.$$

Defining $\Delta = \omega - \omega_0$ as the **Detuning** and considering the situation close to the **resonance condition**: $\Delta << \omega_0$, we obtain

$$\vec{\mu}_0(\omega) \simeq -\frac{e^2}{m\omega_0} \frac{\vec{E}_0}{2\Delta - i\Gamma_{\text{tot}}},$$

thus

$$\alpha_{\text{CL}} = -\frac{e^2}{m\omega_0 \left[2\Delta - i\Gamma_{\text{tot}}\right]},$$

$$\vec{\mu}_0(\omega) = \alpha_{\text{CL}} \vec{E}_0.$$

We now replace $\frac{e^2}{m\omega_0}$ with a term containing Γ, i.e.

$$\frac{e^2}{m\omega_0} = \frac{3}{2}\Gamma\frac{1}{k^3}.$$

The **Polarizability of a Classical Dipole** can thus be written as

$$\alpha_{\text{CL}} = -\frac{3}{2} \frac{1}{k^3} \frac{\Gamma}{2\Delta + i\Gamma_{\text{tot}}}.$$

The complex number represents the presence of damping (radiative and non-radiative).

3 Polarizability of a Two-Level System

We start from the Hamiltonian of a TLS interacting with an applied electric field $\vec{E}$ through the dipole operator $\hat{\vec{d}}$ in the semi-classical theory

$$\hat{H} = \hat{H}_{\text{TLS}} - [\hat{\vec{d}} \cdot \vec{E}],$$

where

$$\hat{H}_{\text{TLS}} = \begin{pmatrix} E_2 & 0 \\ 0 & E_1 \end{pmatrix} = \frac{\hbar\omega_0}{2}\hat{\sigma}_z,$$

where $\omega_0 = \frac{E_2 - E_1}{\hbar}$ is the transition frequency and $\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ is the Pauli matrix. Similarly, we can sue $\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ to write

$$\hat{d} \cdot \vec{E} = \begin{pmatrix} 0 & d_{12}\ E_0\ e^{-i\omega t} \\ d_{21}\ E_0\ e^{-i\omega t} & 0 \end{pmatrix} = \hbar V\ e^{-i\omega t}\hat{\sigma}_x,$$

where V is the **Rabi Frequency**, $V = -d_{12}\frac{E_0}{\hbar}$.

To solve the problem we can use the **Heisenberg Equations of Motion**

$$\dot{\hat{A}} = \frac{i}{\hbar}[\hat{H}, \hat{A}],$$

which we apply to σ_z and σ_x using the **Pauli Matrices**

$$\sigma_+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix},$$

$$\sigma_- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix},$$

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix},$$

$$\sigma_y = \begin{pmatrix} 0 & -i \\ +i & 0 \end{pmatrix}.$$

Also,

$$\sigma_+ = \frac{\sigma_x + i\sigma_y}{2},$$

$$\sigma_- = \frac{\sigma_x - i\sigma_y}{2}.$$

For the oscillating dipole have

$$d_{12}(t) \simeq d_{12}(0)e^{i\omega_0 t}.$$

In the coupling term using the **Rotating Wave Approximation** we can neglect the terms oscillating with fast frequency components, i.e. $\omega + \omega_0$. Hence we get

$$\dot{\hat{\sigma}}_- = (i\Delta - \Gamma_2)\hat{\sigma}_- + \frac{1}{2}iV\hat{\sigma}_z,$$

$$\dot{\hat{\sigma}}_z = -i\Gamma_1)\hat{\sigma}_z + iV[\hat{\sigma}_- - \hat{\sigma}_+]$$

and so on....

Here, Γ_1 and Γ_2 are the **Damping Rates**: Γ_1 is reducing the population and Γ_2 is reducing the dipole moment (coherence). Moreover, the relationship between these two damping terms is

$$\Gamma_2 = \frac{\Gamma_1}{2} + \Gamma_2^*,$$

where Γ_2^* is an additional **Dephasing Rate** for the coherence. Neglecting non-radiative damping, Γ_1 is given by the **Spontaneous Decay**, i.e.,

$$\Gamma_1 = \frac{d_{12}^2\omega_0^3}{3\pi\epsilon_0\hbar c^3},$$

where d_{12} is the amplitude of the dipole moment.

We can write the **Steady State Solution** of the expectation values $<>$ of the operators as

$$< \sigma_- >^{ss} = \frac{V(\Delta - i\Gamma_2)}{2\left[\Delta^2 + \ \Gamma_2^2 + V^2\ \frac{\Gamma_2}{\Gamma_1}\right]},$$

$$< \rho_{22} >^{ss} = \frac{1}{2}\left[1 + < \sigma_z >^{ss}\right] = \frac{V^2\ \Gamma_2}{2\ \Gamma_1\left[\Delta^2 + \ \Gamma_2^2 + V^2\ \frac{\Gamma_2}{\Gamma_1}\right]}.$$

Because $d_{12} < \sigma_- >^{ss}$ represents the expectation value of the dipole moment, the polarizability of a TLS can be written as

$$\alpha_{\text{TLS}} = -\frac{d_{12}\ < \sigma_- >^{ss}}{\frac{1}{2}\ \epsilon_0\ E_0},$$

where the 1/2 terms comes from the fact that

$$E_0 \cos\omega t = \frac{1}{2}\left[e^{i\omega t} + e^{-i\omega t}\right].$$

Hence

$$\alpha_{\text{TLS}} = -\frac{d_{12}^2}{\epsilon_0\hbar}\ \frac{\Delta - i\Gamma_2}{\Delta^2 + \ \Gamma_2^2 + V^2\ \frac{\Gamma_2}{\Gamma_1}}.$$

By replacing d_{12}^2 with Γ_1 we obtain

$$\alpha_{\text{TLS}} = -\frac{3\pi}{k^3} \frac{\Gamma_1[\Delta - i\Gamma_2]}{\Delta^2 + \Gamma_2^2 + V^2 \frac{\Gamma_2}{\Gamma_1}},$$

while for a classical dipole we have

$$\alpha_{CL} = -\frac{6\pi}{k^3} \frac{\Gamma}{2\,\Delta + i\Gamma_{\text{tot}}}.$$

The polarizability of the TLS exhibits parametric coupling with the applied electric field, because of the term V^2 in the denominator. For $V^2 \to +\infty \implies \alpha_{TLS} = 0$, i.e. **under saturation the emission for a TLS is not related to the coherent oscillation of an induced dipole, but to the excited-state population (incoherent emission)**.

4 Coherent/Incoherent Emission of a Two Level System

The **Total Emitted Power** can be written (see lecture notes) as:

$$P_{\text{tot}} = \hbar\,\omega\,\rho_{22}^{ss}\,\Gamma_1 = R_\infty \frac{\frac{I}{I_S}}{1 + \frac{I}{I_S}},$$

where R_∞ represents the emission rate at saturation and I_S is the saturation intensity, which can be related to V^2 (see previous question). The **Coherent Emission** can be related to the field created by the coherence, i.e.

$$E = \alpha_{\text{TLS}}\, E_0 \frac{k^2}{4\pi} \frac{e^{ikr}}{r} (\hat{n} \times \hat{x}) \times \hat{n}$$

where $\hat{n}$ represents the observation direction and $\hat{x}$ is the direction of the oscillating dipole. Therefore, the power related to the coherent part can be written as

$$P_{\text{coh}} = \frac{\sigma_0}{4} \frac{\Gamma_1^2(\Delta^2 + \Gamma_2^2)}{\left[\Delta^2 + \Gamma_2^2 + V^2 \frac{\Gamma_2}{\Gamma_1}\right]^2} I,$$

where I is the **Intensity**, $I = \frac{1}{2}\frac{E_0^2}{Z}$, and σ_0 is the **Scattering Cross-Section**, $\sigma_0 = \frac{3\lambda^2}{2\pi}$, which is related to the polarizability (for $\Delta = 0$ and $V = 0$) through the expression $\sigma_0 = k^4|\alpha_{\text{TLS}}|^2/6\pi$. By introducing the **Saturation** parameter

$$S = \frac{V^2\,\Gamma_2}{(\Delta^2 + \Gamma_2^2)_1} \simeq \frac{I}{I_S},$$

we obtain,

$$P_{\text{tot}} = \frac{\hbar\Gamma_1}{2} \frac{S}{1+S},$$

$$P_{\text{coh}} = \frac{\hbar\Gamma_1}{2} \frac{\Gamma_1}{2\Gamma_2} \frac{S}{(1+S)^2}.$$

The incoherent power is simply the difference between the total and the coherent power,

$$P_{\text{incoh}} = P_{\text{tot}} - P_{\text{coh}} = \frac{\hbar\Gamma_1}{2} \frac{S}{(1+S)^2}\left[S + 1 - \frac{\Gamma_1}{2\Gamma_2}\right].$$

For low excitation, the scattered power is like that of a **classical dipole**, i.e. $P_{\text{tot}} = P_{\text{coh}}$, whereas above saturation the power is incoherent, i.e. $P_{\text{tot}} = P_{\text{incoh}}$.

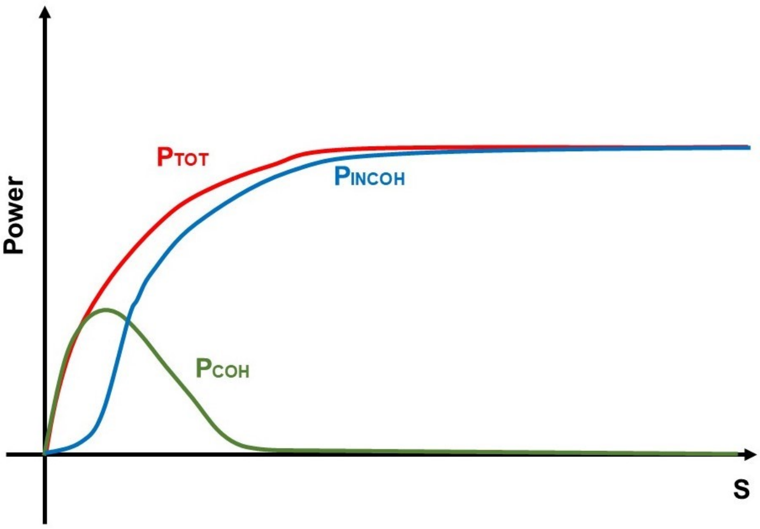

Figure 1: Power emitted by a TLS as a function of the saturation parameter S.

5 References

1. Principles of Nano-Optics (Second edition) by Lukas Novotny
2. Molecular scattering and fluorescence in strongly-confined optical fields by Mario Agio