

Two-level Systems, Part 2:

Dynamics of a Two-level (Atomic or Molecular) System Driven by an Optical field.

Consider a (time-independent) Hamiltonian \hat{H}_0

Let $\phi_{1,2}(x)$ be energy eigenfunction of this Hamiltonian, i.e., $\hat{H}_0\phi_{1,2}(x) = E_{1,2}\phi_{1,2}(x)$

Now, illuminate this atomic/molecular system with a monochromatic light source.

The total Hamiltonian governing the motion of the system is then:

$$\hat{H}(t) = \hat{H}_0 - \hat{\mu}\mathcal{E}_0 \cos(\omega_0 t)$$

Here: $\hat{\mu}$ = electric dipole moment operator

\mathcal{E}_0 = electric field strength

ω_0 = light frequency

Assume the relevant matrix elements of the electric dipole operator are:

$$\int dx \phi_1(x) \hat{\mu} \phi_1(x) = 0 = \int dx \phi_2(x) \hat{\mu} \phi_2(x)$$

$$\int dx \phi_1(x) \hat{\mu} \phi_2(x) = -\mu_0 = \int dx \phi_2(x) \hat{\mu} \phi_1(x)$$

Then, representing the time-dependent wavefunction as a superposition of these two basis functions, as indicated in Eq. [1] above, the time-dependent Schrodinger Eq. is converted into the following matrix form:

$$i\hbar \begin{pmatrix} \dot{c}_1(t) \\ \dot{c}_2(t) \end{pmatrix} = \mathbf{H}(t) \begin{pmatrix} c_1(t) \\ c_2(t) \end{pmatrix} ; \mathbf{H}(t) \equiv \begin{bmatrix} E_1 & \mu_0 \mathcal{E}_0 \cos(\omega_0 t) \\ \mu_0 \mathcal{E}_0 \cos(\omega_0 t) & E_2 \end{bmatrix} \quad [3]$$

We can solve Eq. [3] to a good approximation when the light frequency is high, and nearly resonant with the energy difference E_2-E_1 .

To see how, we first briefly revisit the static TLS problem analyzed above in Eq. [2]. Define:

$$b_1(t) = e^{i\epsilon t/\hbar} c_1(t) ; b_2(t) = e^{-i\epsilon t/\hbar} c_2(t) \quad [4]$$

Substituting into Eq. [2], generates the equivalent equation of motion:

$$i\hbar \begin{pmatrix} \dot{b}_1(t) \\ \dot{b}_2(t) \end{pmatrix} = \mathbf{H}(t) \begin{pmatrix} b_1(t) \\ b_2(t) \end{pmatrix} ; \mathbf{H}(t) \equiv \begin{bmatrix} 0 & \Delta e^{2i\epsilon t/\hbar} \\ \Delta e^{-2i\epsilon t/\hbar} & 0 \end{bmatrix} \quad [2']$$

Note: Eqs. [2] and [2'] are completely equivalent!

Now we turn our attention to the field-driven TLS Hamiltonian, Eq. [3].

Applying the substitution (Eq. [4]), $b_1(t) = e^{iE_1 t/\hbar} c_1(t)$; $b_2(t) = e^{iE_2 t/\hbar} c_2(t)$

generates the following matrix Schrodinger Eq., which is fully equivalent to Eq. [3]:

$$i\hbar \begin{pmatrix} \dot{b}_1(t) \\ \dot{b}_2(t) \end{pmatrix} = \mathbf{H}(t) \begin{pmatrix} b_1(t) \\ b_2(t) \end{pmatrix} \quad ;$$

$$\mathbf{H}(t) \equiv \begin{bmatrix} 0 & \mu_0 \mathcal{E}_0 \cos(\omega_0 t) e^{-i(E_2 - E_1)t/\hbar} \\ \mu_0 \mathcal{E}_0 \cos(\omega_0 t) e^{i(E_2 - E_1)t/\hbar} & 0 \end{bmatrix} \quad [3']$$

A useful approximation to Eq. [3'] can be generated via the following reasoning

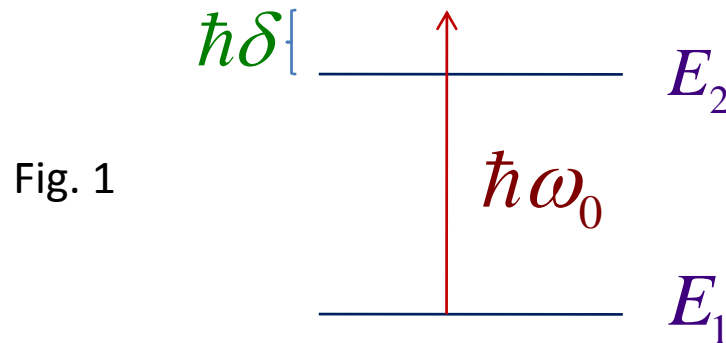
Consider the term:

$$\cos(\omega_0 t) e^{-i(E_2 - E_1)t/\hbar} = \frac{1}{2} \left[e^{i\omega_0 t} + e^{-i\omega_0 t} \right] e^{-i(E_2 - E_1)t/\hbar} \cong \frac{1}{2} e^{i[\omega_0 - (E_2 - E_1)/\hbar]t}$$

The approximation indicated here is justified when the light frequency is large and the detuning frequency (see Fig. 1):

$$\delta \equiv \omega_0 - (E_2 - E_1) / \hbar$$

is small, because then the neglected term above oscillates very rapidly around a mean value of 0, while the retained term oscillates slowly. [Note: This approximation is often termed the **Rotating Wave Approximation=RWA.**]



Assuming the RWA above is valid, then Eq. [3'] is well-approximated by ...

$$i\hbar \begin{pmatrix} \dot{b}_1(t) \\ \dot{b}_2(t) \end{pmatrix} = \mathbf{H}(t) \begin{pmatrix} b_1(t) \\ b_2(t) \end{pmatrix} \quad ;$$

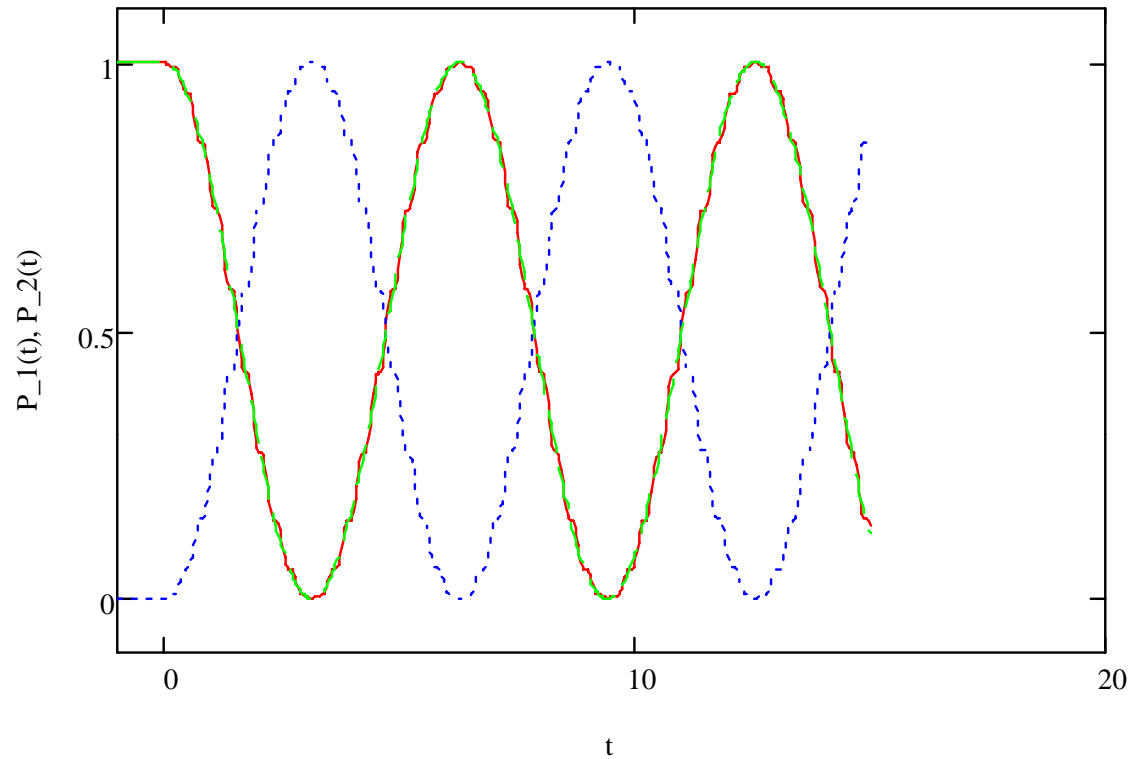
$$\mathbf{H}(t) \equiv \begin{bmatrix} 0 & \frac{\mu_0 \mathcal{E}_0}{2} e^{i\delta t} \\ \frac{\mu_0 \mathcal{E}_0}{2} e^{-i\delta t} & 0 \end{bmatrix} \quad [3'']$$

Note: Eq. [3''] is not exactly equivalent to Eq. [3], but is approximately equivalent to it when the near-resonant, high frequency light source conditions prescribed above obtain.

Assuming Eq. [3''] is approximately valid, we have mapped the driven TLS to the standard, static TLS problem solved above!

Specifically: $\mu_0 \mathcal{E}_0 / 2 \rightarrow \Delta$; $\hbar \delta / 2 \rightarrow \varepsilon$

Illustration of monochromatically driven TLS dynamics:



Plots of $P_1(t)$, $P_2(t)$ based on numerically exact integration of Eq. [3] for parameters noted below. Solid line (red): $P_1(t)$; dotted line (blue): $P_2(t)$. Also plotted via the dashed line is the RWA solution for $P_1(t)$. [Note: Electric field is switched on at $t=0$.]

Parameters: $\mu_0 \mathcal{E}_0 = 1$; $\omega_0 = 10$; $\varepsilon_2 = 10$; $\varepsilon_1 = 0$; $\hbar = 1$