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Interaction of light with matter

When light propagates through matter, the $\vec{E}(r,t)$ field changes the behavior of the matter.

$$i\hbar \frac{\partial}{\partial t} |\Psi_{\text{matter}}\rangle = \hat{H}_0 |\Psi_{\text{matter}}\rangle + \hat{H}' |\Psi_{\text{matter}}\rangle$$

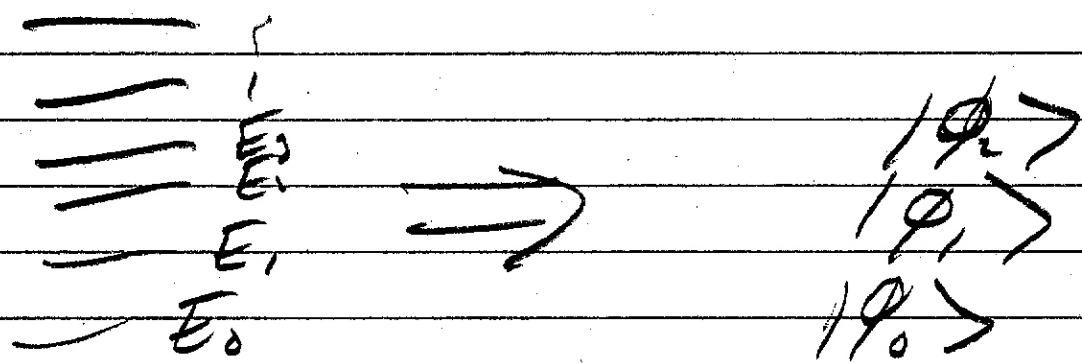
\uparrow
Efield

For now, we are going to treat the light as a classical vector field (not quantum mechanically)

This turns the interaction Hamiltonian into a time dependant operator on the QM state of the matter

The state $|\Psi_{\text{matter}}\rangle$ and \hat{H} Hamiltonian for matter is in general very complex - but we are not going to solve for them - We are just going to assume some solutions exist.

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We are then going to use them as a basis and use time dependant perturbation theory to figure out the behavior of the system

we then have $|\Psi\rangle \Rightarrow \begin{pmatrix} \dots & |\Psi_0\rangle \\ \dots & |\Psi_1\rangle \\ \dots & |\Psi_2\rangle \\ \text{etc.} \end{pmatrix}$

& we have $H_0 = \begin{pmatrix} E_0 & & \\ & E_1 & \\ & & E_2 \\ & & \dots \end{pmatrix}$

& we have $H' = \begin{pmatrix} H'_{11} & H'_{12} & & \\ H'_{21} & H'_{22} & & \\ & & H'_{33} & \\ & & & \dots \end{pmatrix}$
etc.

+ we are going to assume
that the E fields are weak
enough so that time dependent
perturbation theory works
+ H' is linear in the E field.

$$H' = \begin{pmatrix} M_{11} & M_{12} & \\ M_{21} & M_{22} & \\ & M_{33} & \ddots \end{pmatrix} E_0 e^{i\omega t} R(r)$$

$$+ \begin{pmatrix} M_{11}^* & M_{12}^* & \\ M_{21}^* & M_{22}^* & \\ & M_{33}^* & \ddots \end{pmatrix} E_0^* e^{-i\omega t} R^*(r)$$

but we then realize that first
order time dependent perturbation theory
has that nice δ function;

$$W_{m-k} = \frac{2\pi}{\hbar} |H'_{km}|^2 \delta(E_k - E_m - \hbar\omega)$$

+ we realize that the only rates
which are going to be important are
those where $\omega = E_k - E_m$

If we assume there is only one
pair of Energy levels for which
that is true we can ignore the
others & analyse a 2-state system.

$$H = \begin{pmatrix} E_1 & 0 \\ 0 & E_2 \end{pmatrix} \quad |1\rangle, |2\rangle$$

$$+ H' = \begin{pmatrix} M_{11} & M_{12} \\ M_{12}^* & M_{22} \end{pmatrix} E_0 e^{i\omega t} R(r)$$

$$+ \begin{pmatrix} M_{11} & M_{12} \\ M_{12} & M_{22} \end{pmatrix} E_0^* e^{-i\omega t} R^*(r)$$

then we pick the phase of

$$|1\rangle + |2\rangle \text{ to make } M_{12} \text{ real}$$

+ we ignore the effect of $M_{11} + M_{22}$
which does not induce any transitions

$$\text{so } H = \begin{pmatrix} E_1 & 0 \\ 0 & E_2 \end{pmatrix} \quad |1\rangle, |2\rangle$$

$$H' = \begin{pmatrix} 0 & M \\ M & 0 \end{pmatrix} [E_0 e^{i\omega t} R(r) + E_0^* e^{-i\omega t} R^*(r)]$$

Now that we have simplified the problem - we want to take into account the small differences between different atoms in a population, so we use density matrix analysis;

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$$\rho = \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix}$$

$\rho_{11} = C_1 C_1^*$ statistical pop
of level 1

$\rho_{22} = C_2 C_2^*$ " level 2

$\rho_{12} = \rho_{21}^* = C_2 C_1^*$
correlation of
QM phase between
state 1 & state 2
at each atom

Once we have the density matrix
as a function of time, then
we can find any expectation value
averaged over the population

$$|\Psi\rangle = C_1(t)|\phi_1\rangle + C_2(t)|\phi_2\rangle$$

$$i\hbar \frac{\partial}{\partial t} |\Psi_{\text{tot}}\rangle = H |\Psi_{\text{tot}}\rangle$$

$$i\hbar C_m(t) = \sum_n C_n(t) H_{mn}$$

$$\text{so } \frac{\partial}{\partial t} \rho_{nm} = \frac{\partial}{\partial t} \{C_m^*(t) C_n(t)\}$$

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$$\frac{\partial \rho_{nm}}{\partial t} = C_n \frac{\partial c_m^*}{\partial t} + C_n \frac{\partial c_n}{\partial t}$$

+ plugging back in)

$$\frac{\partial \rho_{nm}}{\partial t} = \sum_k \left(\rho_{nk} H_{km} - H_{nk} \rho_{km} \right)$$

applying this to our 2-state system,

$$\frac{d\rho_{11}}{dt} = -\frac{i}{\hbar} \left(H'_{21} \rho_{11} + E_2 \rho_{21} - E_1 \rho_{21} - \rho_{21} H'_{11} \right)$$

$$= -\frac{i}{\hbar} \left[H'_{21} (\rho_{11} - \rho_{21}) + (E_2 - E_1) \rho_{21} \right]$$

or $\frac{d(\rho_{21})}{dt} = -i \omega \rho_{21} + i \frac{\mu}{\hbar} [E(t, r) (A_1 - A_2)]$

$$+ \frac{d(\rho_{21}^*)}{dt} = -i \frac{\mu}{\hbar} E(t, r) [A_1 - \rho_{21}^*]$$

using $\rho_{11} + \rho_{22} = 1$

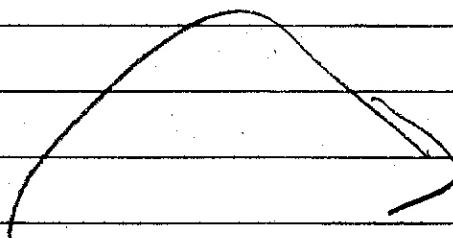
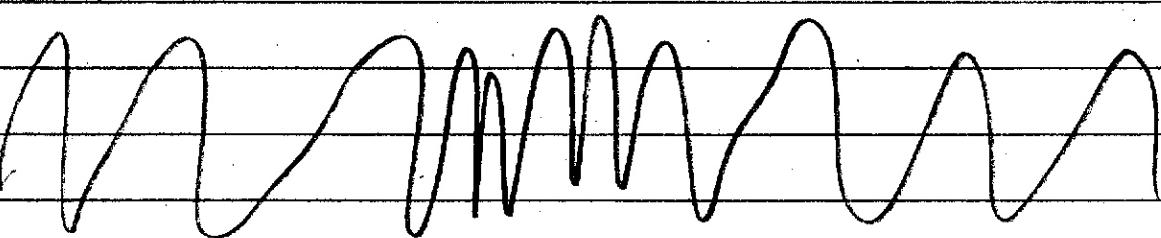
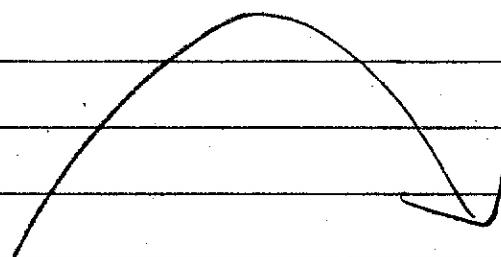
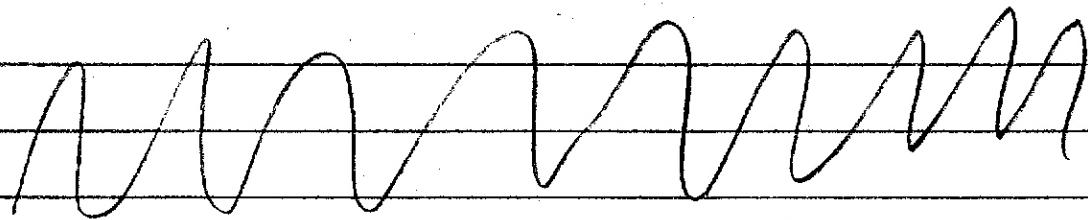
$$\frac{d}{dt} (\rho_{11} - \rho_{22}) = 2i \frac{M}{\hbar} E(t, \nu) (\rho_{21} - \rho_{12}^*)$$

most of which we did a couple of weeks ago.

This is a pair of differential equations for the two independent terms of the density matrix.

- This model, however, does not allow for any net absorption - because there is nowhere for the energy to go. Physically, energy would be transferred to a wide number of other mechanisms by thermal processes. These processes are different for each atom in the ensemble. In addition, there may be random elastic processes which change the progression of the state of the system differently for each atom (member of the ensemble)

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the net effect is that after a period of time, phase correlation between different members of an ensemble are destroyed.

- If the rate at which phase correlation dissipation is proportional to the amount of phase correlation,

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Then the off diagonal terms should have a d^ying exponential behavior in the absence of perturbation. We can get that behavior by putting in an additional term

$$\frac{dP_{21}}{dt} = -\omega_0 P_{21} + i \frac{\mu}{\hbar} (\rho_{11} - \rho_{22}) E(t) - \frac{P_{21}}{T_2}$$

T_2 is called the relaxation time of the population (relaxation)

This still does not give us a mechanism for releasing energy, however, so we need to model a mechanism which returns the population to an equilibrium value. Again, if the rate of return to equilibrium is proportional to the deviation, we can add an inelastic relaxation time

$$\frac{d}{dt} (\rho_{11} - \rho_{22}) = \frac{2i\mu E(t)}{\hbar} (\rho_{21} - \rho_{21}^*)$$

$$- \frac{(\rho_{11} - \rho_{22}) - (\rho_{11} - \rho_{22})_0}{\zeta}$$

This term includes all of the other processes we have left out

- spontaneous emission
- excitations from other states
- other perturbations
- collisions

it can even represent a turnover in the population with a reservoir population

→ we now want to find the steady state solution to the above equations

However, the terms $\rho_{12} + \rho_{21}$ will not have a zero time derivative, they will oscillate at the driving frequency ω (with a phase shift) — otherwise there won't be any expectation values oscillating)

so let's take

$$\rho_{21} = \delta_{21} e^{-i\omega t}$$

to separate out the time dependency
+ we will take $\frac{d}{dt}(\rho_{11} - \rho_{22}) = 0$
as well

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Solving for $\sigma_{21} + (\rho_{11} - \rho_{22})$
 we get

$$(\rho_{11} - \rho_{22}) = (\rho_{11} - \rho_{22})_0 \frac{1 + (\omega - \omega_0)^2 T_2^2}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega^2 T_2 \epsilon}$$

where $\Omega = \mu E_0 / \hbar \hbar$

$$+ I_m \{\sigma_{21}\} = \frac{\Omega T_2 (\rho_{11} - \rho_{22})_0}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega^2 T_2 \epsilon}$$

$$R_c \{\sigma_{21}\} = \frac{(\omega_0 - \omega)^2 \Omega (\rho_{11} - \rho_{22})_0}{1 + (\omega - \omega_0)^2 T_2^2 + 4\Omega^2 T_2 \epsilon}$$

Now that we have a solution for the density matrix of the QM system, we can find any average expectation value over the population

the question is, what operator corresponds to the observable polarization P^3 ?

P must be a two by two matrix

$$\begin{pmatrix} P_{11} & P_{12} \\ P_{21} & P_{22} \end{pmatrix}$$

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+ if we ignore any possible DC polarization,

$$\mathbf{P} \Rightarrow \begin{pmatrix} 0 & P_{12} \\ P_{21}, 0 \end{pmatrix}$$

+ we must have $P_{12} = P_{21}^*$

$$\mathbf{P} = K \begin{pmatrix} 0 & \mu \\ \mu, 0 \end{pmatrix}$$

+ to get the unit $\text{V}^{+ \text{energy}}$ current, $K = N$
the density of atoms per unit volume

we now have $\langle \vec{P} \rangle = N \langle \mu \rangle$

$$\langle \mu(t) \rangle = \text{tr}(\rho \mu)$$

$$= \mu (\sigma_{11} e^{i\omega t} + \sigma_{21} e^{-i\omega t})$$

$$\langle \mu(t) \rangle = 2 \mu R_c \sigma_{21}(t) \cos \omega t$$

$$+ \text{Im} \sigma_{21}(t) \sin \omega t$$

$$\text{so } \vec{P} = \frac{\mu^2 \Delta N_1 T_2}{\pi} E_0 \left[\frac{\sin \omega t + (w_0 - \omega) T_2 \cos \omega t}{1 + (\omega - w_0)^2 T_2^2 + 4 \beta T_2 \varepsilon} \right]$$