ELECTROMAGNETIC RESPONSE OF MEDIUM: SUSCEPTIBILITY

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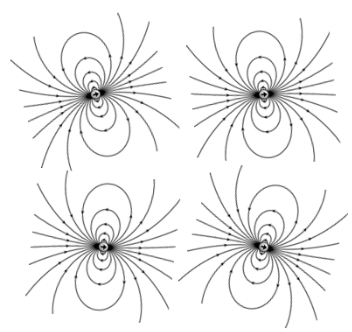
Polarization Equation

$$\frac{d^2x}{dt^2}+\gamma\frac{dx}{dt}+\omega_0^2x=-\frac{e}{m}E_x$$

$$N\langle\mu\rangle=p\Rightarrow$$

$$\frac{d^2p}{dt^2}+\left(\gamma+\frac{2}{T_2}\right)\frac{dp}{dt}+\omega_0^2p=N\frac{e^2}{m}E_x$$



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Excitation

- Consider an external signal $E_x = E_0 e^{j\omega t}$

$$\frac{d^2 p}{dt^2} + \left(\gamma + \frac{2}{T_2}\right)\frac{dp}{dt} + \omega_0^2 p = N\frac{e^2}{m}E_x$$

Transient Solution:

$$\frac{d^2 p}{dt^2} + \left(\gamma + \frac{2}{T_2}\right)\frac{dp}{dt} + \omega_0^2 p = 0$$

$$p_{\text{trans}} = p_i e^{-\left(\frac{\gamma}{2}+\frac{1}{T_2}\right)t} e^{j\sqrt{\omega_0^2 - \left(\frac{\gamma}{2}+\frac{1}{T_2}\right)^2}\, t}$$

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Excitation

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$$\frac{d^2 p}{dt^2} + \left(\gamma + \frac{2}{T_2}\right)\frac{dp}{dt} + \omega_0^2 p = N\frac{e^2}{m}E_x$$

Steady-state solution:

$$p = p_0 e^{j\omega t}$$

$$-\omega^2 p_0 e^{j\omega t} + \left(\gamma + \frac{2}{T_2}\right) j\omega p_0 e^{j\omega t} + \omega_0^2 p_0 e^{j\omega t} = N\frac{e^2}{m}E_0 e^{j\omega t}$$

$$p_0(\omega) = \frac{N\frac{e^2}{m}E_0}{j\omega\left(\gamma + \frac{2}{T_2}\right) + \left(\omega_0^2 - \omega^2\right)}$$

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Total Solution

$$p = p_i e^{-\left(\frac{\gamma}{2}+\frac{1}{T_2}\right)t} e^{j\sqrt{\omega_0^2-\left(\frac{\gamma}{2}+\frac{1}{T_2}\right)^2}\,t} + \frac{N\frac{e^2}{m}E_0}{j\omega\left(\gamma+\frac{2}{T_2}\right)+\left(\omega_0^2-\omega^2\right)} e^{j\omega t}$$

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Response

Excitation:

$$E_x(t) = \mathrm{Re}\left[\tilde{E}_x e^{j\omega t}\right] = \frac{1}{2}\left[\tilde{E}_x e^{j\omega t} + \tilde{E}_x^* e^{-j\omega t}\right]$$

$\tilde{E}_x$: Phasor → $\tilde{E}_x = |\tilde{E}_x| e^{j\phi}$

$$E_x(t) = \mathrm{Re}\left[|\tilde{E}_x| e^{j(\omega t+\phi)}\right] = |\tilde{E}_x| \cos(\omega t+\phi)$$

Steady-state response:

$$p_x(t) = \mathrm{Re}\left[\tilde{P}_x e^{j\omega t}\right] = \frac{1}{2}\left[\tilde{P}_x e^{j\omega t} + \tilde{P}_x^* e^{-j\omega t}\right]$$

In a linear system

$$\tilde{E}_x e^{j\omega t} \leftrightarrow \tilde{P}_x e^{j\omega t} \qquad \tilde{E}_x e^{-j\omega t} \leftrightarrow \tilde{P}_x e^{-j\omega t}$$

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Equation of Motion

$$\frac{d^2 p_x(t)}{dt^2} + \left(\gamma + \frac{2}{T_2}\right)\frac{dp_x(t)}{dt} + \omega_a^2 p_x(t) = N\frac{e^2}{m}E_x(t)$$

$$E_x(t) = \tilde{E}_x e^{j\omega t} \qquad p_x(t) = \tilde{P}_x e^{j\omega t}$$

$$\left[-\omega^2 + j\omega\left(\gamma + \frac{2}{T_2}\right) + \omega_a^2\right]\tilde{P}_x = N\frac{e^2}{m}\tilde{E}_x$$

Transfer function: $$\frac{\tilde{P}_x}{\tilde{E}_x} = \frac{Ne^2}{m}\frac{1}{\omega_a^2 - \omega^2 + j\omega\left(\gamma + \frac{2}{T_2}\right)}$$

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Susceptibility

$$\tilde{D} = \varepsilon_0 \tilde{E} + \tilde{P}$$

$$\tilde{P}(\omega) = \tilde{\chi}(\omega)\varepsilon_0 \tilde{E}(\omega) \Rightarrow \tilde{\chi}(\omega) = \frac{\tilde{P}(\omega)}{\varepsilon_0 \tilde{E}(\omega)}$$

$$\tilde{D} = \varepsilon_0\left[1 + \tilde{\chi}\right]\tilde{E} = \tilde{\varepsilon}\tilde{E}$$

$$\tilde{\varepsilon}(\omega) = \varepsilon_0\left[1 + \tilde{\chi}\right]$$

- For anisotropic medium, $\tilde{\chi}$ becomes a tensor quantity.
  For simplicity, now we will consider $\tilde{\chi}$ a scalar.
- $\tilde{\chi}$ is very important in calculating laser gain, phase shift, and many other properties.

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Background

$$\tilde{D} = \varepsilon_0 \tilde{E} + \tilde{P}_b + \tilde{P}$$

$$\tilde{P}_b = \tilde{\chi}_b \varepsilon_0 \tilde{E} \Rightarrow \varepsilon_b = \varepsilon_0 \left(1 + \tilde{\chi}_b\right)$$

$$\tilde{D} = \varepsilon_b \tilde{E} + \tilde{P}$$

Often we are in a medium with the atoms providing gain, and the field is on resonance with the transition. There are background atoms (for example, crystalline host in a solid-state laser) which has a background dielectric constant much larger than that of the active atoms. Thus we can write for a propagating wave $e^{j(\omega t - kz)}$ and a background dielectric constant ε_b.

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Susceptibility

$$\tilde{\chi}(\omega) = \frac{\tilde{P}(\omega)}{\varepsilon \tilde{E}(\omega)} = \frac{Ne^2}{m\varepsilon} \frac{1}{\omega_a^2 - \omega^2 + j\omega\Delta\omega_a}$$

$$\Delta\omega_a = \gamma + \frac{2}{T_2}$$

$\Delta\omega_a$: Linewidth (FWHM) of the atomic resonance.

$$\Delta\omega_a \ll \omega_a$$

$$\omega \approx \omega_a$$

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Susceptibility

$$\tilde{\chi}(\omega)=\frac{Ne^2}{m\varepsilon}\frac{1}{\omega_a^2-\omega^2+j\omega\Delta\omega_a}=\frac{Ne^2}{m\varepsilon}\frac{1}{(\omega_a-\omega)(\omega_a+\omega)+j\omega\Delta\omega_a}$$

$$=\frac{Ne^2}{m\varepsilon}\frac{1}{2\omega(\omega_a-\omega)+j\omega\Delta\omega_a}=\frac{Ne^2}{m\varepsilon}\frac{1}{j\omega\Delta\omega_a\left[1+\frac{2(\omega_a-\omega)}{j\Delta\omega_a}\right]}$$

$$\tilde{\chi}(\omega)=-j\frac{Ne^2}{m\omega_a\varepsilon\Delta\omega_a}\frac{1}{1+2j\frac{\omega-\omega_a}{\Delta\omega_a}}$$

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Lorentzian Lineshape

$$\tilde{\chi}(\omega)=-j\frac{Ne^2}{m\omega_a\varepsilon\Delta\omega_a}\frac{1}{1+2j\frac{\omega-\omega_a}{\Delta\omega_a}}$$

Let us take $\Delta x=2\frac{\omega-\omega_a}{\Delta\omega_a}$ and $\chi_0=\frac{Ne^2}{m\omega_a\varepsilon\Delta\omega_a}$

$$\tilde{\chi}(\omega)=-j\chi_0\frac{1}{1+2j\frac{\omega-\omega_a}{\Delta\omega_a}}=-j\chi_0\frac{1}{1+j\Delta x}$$

The complex lorentzian lineshape is simply the Fourier transform in frequency space of the exponential time decay of the polarization $p_x(t)$.

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Real and Imaginary Parts

$$\tilde{\chi}(\omega) = \chi'(\omega) + j\chi''(\omega)$$

$$\chi''(\omega) = -\chi_0 \frac{1}{1 + \left[2\dfrac{\omega - \omega_a}{\Delta\omega_a}\right]^2} = -\chi_0 \frac{1}{1 + \Delta x^2}$$

$$\chi'(\omega) = -\chi_0 \frac{2\dfrac{\omega - \omega_a}{\Delta\omega_a}}{1 + \left[2\dfrac{\omega - \omega_a}{\Delta\omega_a}\right]^2} = -\chi_0 \frac{\Delta x}{1 + \Delta x^2}$$

$$\tilde{\chi}(\omega) = -\chi_0 \left[\frac{\Delta x}{1 + \Delta x^2} + j\frac{1}{1 + \Delta x^2}\right]$$

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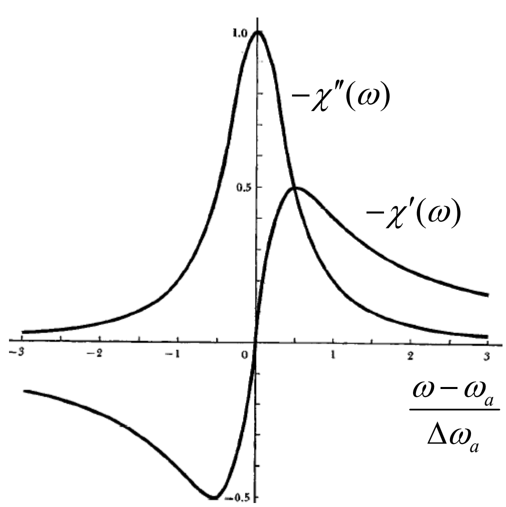
Real and Imaginary Parts



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Linewidth

- Transition linewidth is proportional to $\Delta\omega_a$ and χ_0 is proportional to $1/\Delta\omega_a$ at resonance.
- The peak of the susceptibility becomes larger as the linewidth becomes smaller.

$$\Delta\omega_a = \left(\gamma + \frac{2}{T_2}\right) = \gamma_{\text{rad}} + \gamma_{\text{nr}} + \frac{2}{T_2}$$

- In addition to radiative and non-radiative decay rates, the linewidth is broadened by dephasing collisions. Thus if one increases the dephasing collision rate, the linewidth broadens and the peak decreases! We can easily see this in gases where we can control the collision rate through pressure.

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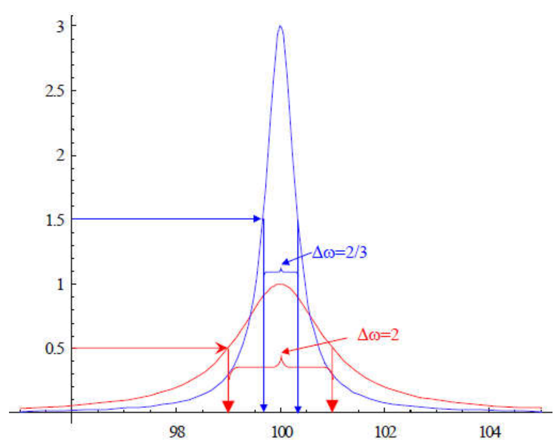
Variation with Linewidth



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CONVERSION TO REAL ATOMIC TRANSITIONS

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Radiative Decay Rate

$$\chi_0 = \frac{Ne^2}{m\omega_a \varepsilon \Delta\omega_a}, \qquad \gamma_{\text{rad}} = \frac{e^2\omega_a^2}{6\pi\varepsilon m c^3}$$

$$\chi_0 = \frac{3}{4\pi^2}\frac{N\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a}$$

- The atomic and electromagnetic constants appearing, e.g., charge *e*, mass *m*, and the dielectric constant ε drop out. Now χ_0 depends on transition wavelength, the density of oscillators, and the radiative decay rate.

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Population Difference

$$\chi_0 = \frac{3}{4\pi^2}\frac{N\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a} \Rightarrow \frac{3}{4\pi^2}\frac{\Delta N_{12}\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a} = \frac{3}{4\pi^2}\frac{\left[N_1 - N_2\right]\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a}$$

- N_1 and N_2 are the level populations of the lower and upper energy levels.

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Quantum Susceptibility

$$\tilde{\chi}(\omega) = -j\frac{3}{4\pi^2}\frac{\Delta N\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a}\frac{1}{1+2j\dfrac{\omega-\omega_a}{\Delta\omega_a}}$$

- This expression is essentially a quantum-mechanically correct expression.

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Quantum Polarization Equation

$$\frac{d^2 p_x(t)}{dt^2} + \Delta\omega_a \frac{dp_x(t)}{dt} + \omega_a^2 p_x(t) = N\frac{e^2}{m}E_x(t)$$

$$N\frac{e^2}{m} = \frac{3}{4\pi^2}\omega_a \varepsilon \lambda^3 \gamma_{\text{rad}} \Delta N$$

$$\frac{d^2 p_x(t)}{dt^2} + \Delta\omega_a \frac{dp_x(t)}{dt} + \omega_a^2 p_x(t) = \frac{3}{4\pi^2}\omega_a \varepsilon \lambda^3 \gamma_{\text{rad}} \Delta N(t) E_x(t)$$

- Now, $\Delta N(t)$ can be a function of time in contrast to a fixed N in classical model. This makes the quantum equation nonlinear.

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Limits

$$\frac{d^2 p_x(t)}{dt^2} + \Delta\omega_a \frac{dp_x(t)}{dt} + \omega_a^2 p_x(t) = \frac{3}{4\pi^2}\omega_a \varepsilon \lambda^3 \gamma_{\text{rad}} \Delta N(t) E_x(t)$$

- Slowly varying $\Delta N(t) \rightarrow$ rate-equation limit, linear equation.
- Strong applied signal $\rightarrow$ nonlinear polarization equation, Rabi flopping behavior.

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ELECTRIC-DIPOLE TRANSITIONS

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Rates

- It is important to remember that rates add.
- Remember the old lawn mowing question: If *A* can mow a lawn in 30 minutes and his brother *B* can mow it in 20 minutes, how long does it take them together to mow the lawn?
- What we need to calculate is lawns per minute.
- *A*'s rate = 1/30 lawns minute; *B*'s rate = 1/20 lawns per minute. Since they are working together the lawn gets mowed at the sum of the rates. Thus the total rate is 1/30+1/20=5/60=1/12 lawns per minute. This means that one lawn gets done in 12 minutes!!

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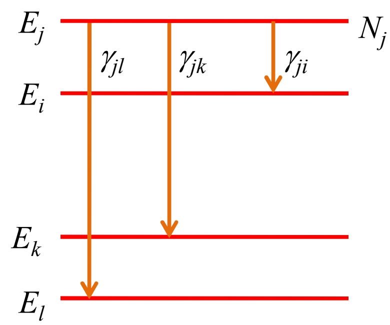
Rates

$$\frac{dN_j}{dt} = -\left(\gamma_{ji} + \gamma_{jk} + \gamma_{jl}\right) N_j$$
$$= -\gamma_j N_j = -N_j / \tau_j$$



- Generalization:

$$\frac{dN_j}{dt} = -\sum \gamma_{ji} N_j$$
$$= -\gamma_j N_j = -N_j / \tau_j$$
$$N_j = N_{j0} e^{-\gamma_j t} = N_{j0} e^{-t/\tau_j}$$

- For any transition: $\gamma_{ji} = \gamma_{ji,\text{rad}} + \gamma_{ji,\text{nr}}$

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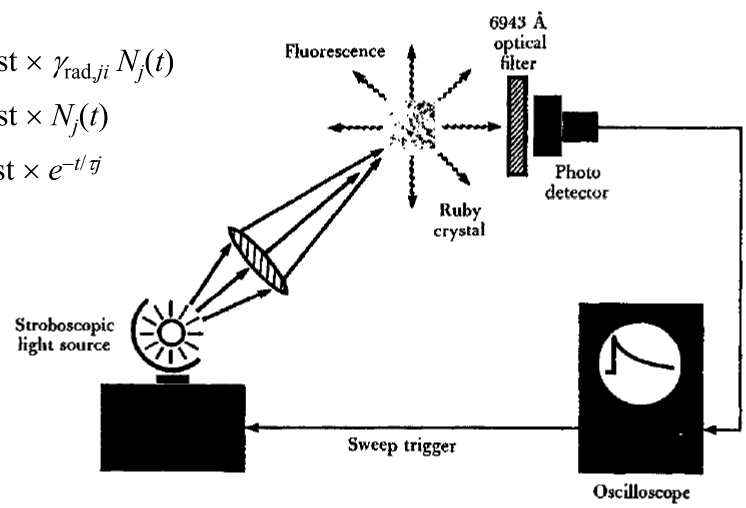
Fluorescence Lifetime Measurement

- $I_{\text{fl}}(t) = \text{const} \times \gamma_{\text{rad},ji} N_j(t)$
- $I_{\text{fl}}(t) = \text{const} \times N_j(t)$

$$= \text{const} \times e^{-t/\tau_j}$$



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Radiative Rates

- Both the stimulated and spontaneous emission rates are proportional to this dipole moment squared.

$$\vec{\mu}_{ji} = \int d^3\vec{r}\,\psi_j^*(\vec{r})(-e\vec{r})\psi_i(\vec{r})$$

$$\gamma_{ji,\text{rad}} = A_{ji} = \frac{8\pi^2}{\varepsilon\hbar\lambda^3}\left|\int d^3\vec{r}\,\psi_j^*(\vec{r})(-e\vec{r})\psi_i(\vec{r})\right|^2$$

Wavefunctions

- The scalar function ψ_j is the solution to Schrdinger's equation. In the case of the Hydrogen atom we can get analytic solutions.
- The Eigenvalues are the atom's energy levels.
- $|\psi_j(\mathbf{r})|^2$ is the probability per unit volume of finding the electron at the coordinate **r**.
- When we write the dipole moment as on the previous slide this is an integral over all space. This is called an expectation value which is like an average.