1 The Oscillator model, Homogeneous Broadening

The Einstein coefficients were a first phenomelogical approach to the interaction between radiation — 2 level systems — and matter. The microscopic approach is to calculate the response of electrons to the applied alternating electric field of the light. This should really be done quantum-mechanically. A purely classical model can however give a good physical understanding.

The electron attachment to the atom or molecule is represented by a spring constant K. The motion is damped. The damping term (proportional to the velocity) is represented by a characteristic time T_2, of which the physical significance will become clear later. One solves the equation of motion of the electron under the light field. The oscillating field induces a periodic motion $x(t)$ of the field. The oscillating dipole $ex(t)$ radiates. *Absorption* results from the destructive interference between the applied field, and the field radiated by the dipoles, when this field is 180° out of phase with the applied field.

The equation of motion of the electron is:

$$\begin{aligned}\frac{d^2x}{dt^2}+\frac{2}{T_2}\frac{dx}{dt}+\frac{K}{m}x &= \frac{e}{m}E(t)\\ \frac{d^2x}{dt^2}+\frac{2}{T_2}\frac{dx}{dt}+\omega_0^2 x &= \frac{e}{m}E(t).\end{aligned} \tag{1}$$

Using for the electric field $E(t)=\frac{1}{2}\mathcal{E}_0 e^{i\omega t}$, we search for a solution of the form $x(t)=\frac{1}{2}Xe^{i\omega t}$. After substitution, one finds:

$$X=\frac{e\mathcal{E}_0}{2m\omega_0[\Delta\omega T_2+i]} \tag{2}$$

where $\Delta\omega=\omega_0-\omega$. The polarization is:

$$P=\frac{1}{2}\tilde{\mathcal{P}}e^{i\omega t}=\frac{1}{2}NeXe^{i\omega t} \tag{3}$$

The polarization amplitude is:

$$\tilde{\mathcal{P}}=\frac{Ne^2T_2\mathcal{E}_0}{2m\omega_0}\frac{i-\Delta\omega T_2}{1+\Delta\omega^2T_2^2}=\epsilon_0\left(\chi'-i\chi''\right)\mathcal{E}. \tag{4}$$

where we have defined the real and imaginary parts of the susceptibility.

Let us now try to make a connection to the absorption defined previously through the rate equations. The absorption coefficient is $\epsilon_0\chi''=N\sigma$. This makes the first connection between the dipole moment and the cross section. Now we know that the frequency dependence of the cross section is:

$$\sigma=\frac{\sigma_0}{1+\Delta\omega^2T_2^2}. \tag{5}$$

The implication is that the saturation energy density and the saturation intensity increase off resonance:

$$W_s = \frac{h\nu}{2\sigma} = \frac{h\nu}{2\sigma_0}(1+\Delta\omega^2 T_2^2) = W_{s0}(1+\Delta\omega^2 T_2^2). \tag{6}$$

Substituting in the rate equation:

$$\frac{d\Delta N}{dt} = \frac{I}{W_{s0}(1+\Delta\omega^2 T_2^2)}\Delta N - \frac{\Delta N - \Delta N_0}{T_1}. \tag{7}$$

Solving for the steady state:

$$\Delta N_{eq} = \frac{1+\Delta\omega^2 T_2^2}{1+\Delta\omega^2 T_2^2 + \frac{I}{I_s}}. \tag{8}$$

This is the density of atoms of atoms to be inserted in the definition of the susceptibility. The result leads to:

$$\alpha_r = \frac{\alpha_0}{1+\Delta\omega^2 T_2^2 + \frac{I}{I_s}} \tag{9}$$

$$\alpha_i = \frac{\alpha_0 \Delta\omega T_2}{1+\Delta\omega^2 T_2^2 + \frac{I}{I_s}} \tag{10}$$

This expression can be put in many different forms,depending which kind of dependence one wants to emphasize. Eqs. (9,10) are the imaginary and real parts of the complex function:

$$\frac{\frac{1}{2}\frac{\alpha_0}{1+\frac{I}{I_s}}}{\Delta\omega\frac{T_2}{\sqrt{1+\frac{I}{I_s}}}+i}. \tag{11}$$

As an intense ($I > I_s$) beam is scanned across the line, the saturation intensity increases away from the line. Since the line is saturated more in the center than in the wings, it appears broadened. The linewidth measured with an intense beam would be:

$$\Delta\omega_{FWHM}(I) = \frac{2}{T_2}\sqrt{1+\frac{I}{I_s}}. \tag{12}$$

The lineshape can be understood by rewriting Eqs (9,10) in the form:

$$\alpha_r = \frac{\frac{\alpha_0}{1+\frac{I}{I_s}}}{1+\frac{\Delta\omega^2 T_2^2}{1+\frac{I}{I_s}}} \tag{13}$$

$$\alpha_i = \frac{\frac{\alpha_0 \Delta\omega T_2}{1+\frac{I}{I_s}}}{1+\frac{\Delta\omega^2 T_2^2}{1+\frac{I}{I_s}}} \tag{14}$$

It is a different situation if a strong pulse is applied for instance at resonance, to saturate the transition, and a probe pulse is used to measure the linewidth. In that case, the Lorentzian lineshape is unchanged.

1.1 Propagation

We can always write for the absorption coefficient:

$$\alpha = \frac{\alpha_{\mathrm{off-resonance}}}{1 + \frac{I}{I_{s2}}} = \frac{\frac{\alpha_0}{1+\Delta\omega^2 T_2^2}}{1 + \frac{I}{I_s(1+\Delta\omega^2 T_2^2)}}. \tag{15}$$

Therefore, for propagation through thick gain/absorber media:

$$\frac{I - I_0}{I_{s2}} + \ln\frac{I}{I_0} = \alpha z \tag{16}$$

whether on resonance or not.

2 Inhomogeneous Broadening

Instead of a two-level system at one frequency ω_0, we have a frequency distribution g of two level systems. Therefore the absorption coefficient takes the form:

$$\alpha = \alpha_0 \int_{-\infty}^{\infty} \frac{g(\omega_0) d\omega_0)}{1 + \Delta\omega^2 T_2^2 + \frac{I}{I_s}}. \tag{17}$$