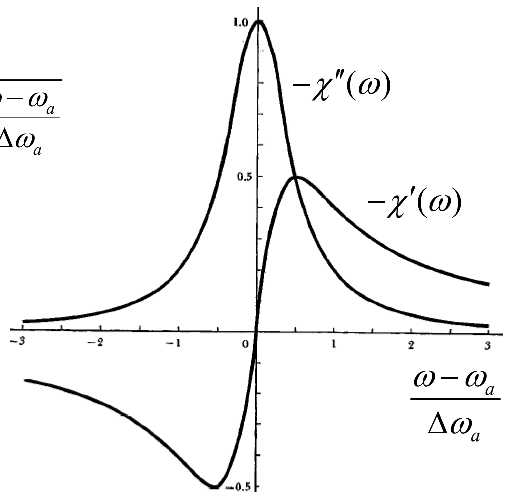
LINE-BROADENING

Susceptibility and Linewidth

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

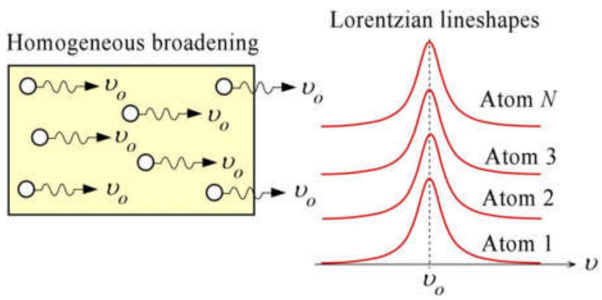
- Transition linewidth is proportional to $\Delta\omega_a$ and χ_0 is proportional to $1/\Delta\omega_a$ at resonance.
- The peak of the susceptibility becomes larger as the linewidth becomes smaller.



Homogeneous Broadening

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

$$\Delta\omega_a = \gamma_i + \gamma_j + \frac{2}{T_{2,ij}}$$



- Energy decay and dephasing act on all the atoms in a collection in the same way.
- Lineshape is Lorentzian for homogeneous broadening.
- Examples of homogeneous broadening are natural or lifetime broadening, and collisional or pressure broadening. In these cases each system is affected "on average" in the same way (e.g. by the collisions due to the pressure).

Lifetime Broadening

- If dephasing effect is absent:

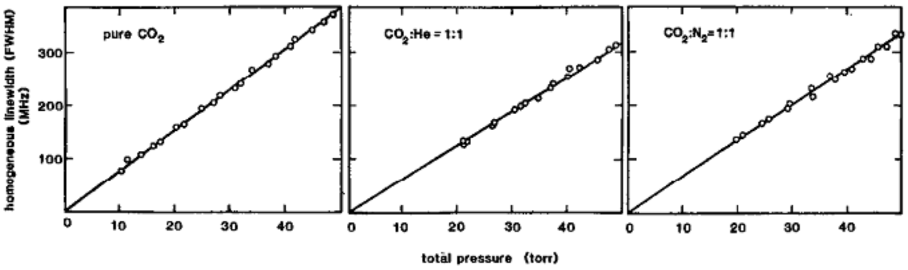
$$\Delta\omega_a = \gamma = \gamma_{\text{rad}} + \gamma_{\text{nr}}$$

- Purely radiative broadening

$$\Delta\omega_a = \gamma_{\text{rad}}$$

- Examples of lifetime broadening are found in very low-pressure gases.
- In a gas, $\Delta\omega_a / 2\pi$ can range from a few MHz to tens of MHz. In solids $\Delta\omega_a / 2\pi$ can be much greater due to interactions with the phonons.

Pressure Broadening



$$\Delta\omega_a = A + BP$$

A, *B*: Constants
P: Pressure

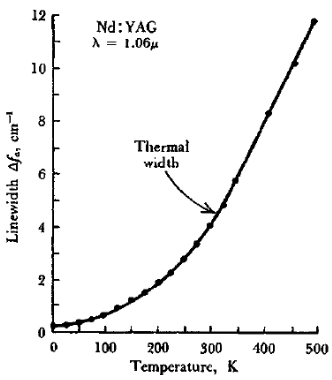
In case of a mixture of collision partners:

$$\Delta\omega_a = A + B_{He}P_{He} + B_{N_2}P_{N_2} + B_{CO_2}P_{CO_2}$$

5

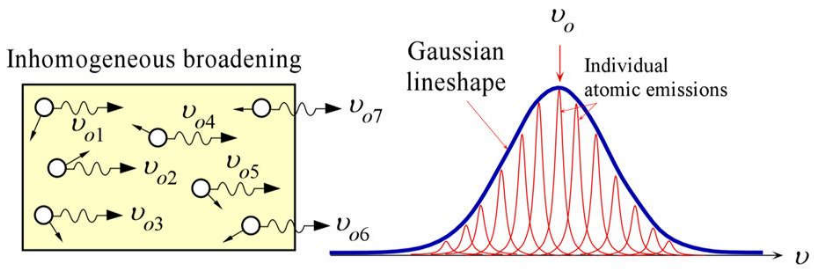
Phonon Broadening

- Phonon broadening refers to a rapid and random frequency modulation of the instantaneous atomic transition frequency for an atom in a solid caused by high-frequency lattice vibrations.
- Phonon broadening does not depend on atomic density *N*, however, depends strongly on lattice temperature.



6

Inhomogeneous Broadening



- Groups of identical atoms see different environment.
- Lineshape is Gaussian for inhomogeneous broadening.
- The most frequent situation in solid state systems where the fluctuation is different for each system (**inhomogeneous broadening**) is when because of the presence of dopants, the local electric field is different for each emitter, and so the Stark effect changes the energy levels in an inhomogeneous way.

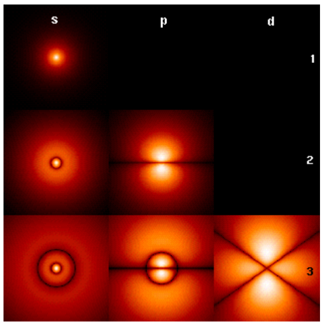
7

POLARIZATION

8

Hydrogen Wavefunctions

n	l	m_l		$\psi_{nlm_l}(r,\theta,\phi)$
1	0	0	$1s$	$\frac{1}{\sqrt{\pi}a_0^{3/2}}e^{-r/a_0}$
2	0	0	$2s$	$\frac{1}{4\sqrt{2\pi}a_0^{3/2}}\left[2-\frac{r}{a_0}\right]e^{-r/2a_0}$
2	1	0	$2p$	$\frac{1}{4\sqrt{2\pi}a_0^{3/2}}\frac{r}{a_0}e^{-r/2a_0}\cos\theta$
2	1	± 1	$2p$	$\frac{1}{8\sqrt{\pi}a_0^{3/2}}\frac{r}{a_0}e^{-r/2a_0}\sin\theta\, e^{\pm j\phi}$



$$a_0 = \frac{\hbar^2}{me^2} = 0.0529\,\text{nm} = \text{first Bohr radius}$$

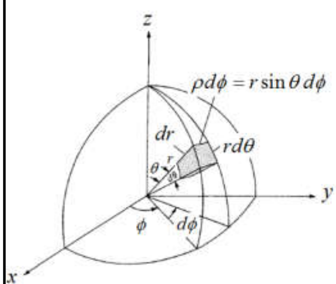
Dipole Moment

Consider a linearly polarized field in z-direction. Let's evaluate the z component of the dipole moment, μ_z, for Hydrogen atom.

$$\mu_{zji} = \int d^3r\, \psi_j^*(\vec{r})(-ez)\psi_i(\vec{r})$$

$$z = r\cos\theta$$

$$d^3r = r^2\, dr \sin\theta\, d\theta\, d\phi$$



$$\mu_{z,1s\to 2p}$$

$$= \int d^3\vec{r}\, \psi_{1s}^*(r)\, z\, \psi_{2p}(\vec{r})$$

$$= \int_0^\infty dr\, r^2 \frac{r}{\pi a_0^4} e^{-\frac{3r}{2a_0}}$$

$$\times r\int_0^{2\pi} d\phi \int_0^{\pi} d\theta \sin\theta\cos\theta \begin{matrix}\frac{1}{4\sqrt{2}}\cos\theta \\ \frac{1}{8}\sin\theta\, e^{\pm j\phi}\end{matrix}$$

Selection Rules

- Here we note that the integrals over θ give us the result we are looking for.

$$\int_0^{\pi} \cos\theta \sin^2\theta \, d\theta = 0$$

$$\int_0^{\pi} \cos^2\theta \sin\theta \, d\theta = \frac{2}{3}$$

- Thus the only state that participates is the $l = 1$, $m = 0$ term ($\cos\theta$ term in the 2p wavefunction).

Tensor Susceptibility

$$\vec{\mathrm{P}}(\omega) = \varepsilon \,\overline{\overline{\chi}}(\omega)\, \vec{\mathrm{E}}(\omega)$$

- In the general case, the tensor is a 3 by 3 matrix and may not be simple. This is true if the material is composed of different atoms/molecules in different directions.

$$\tilde{P}_x(\omega) = \varepsilon\, \tilde{\chi}_{xx}(\omega) \tilde{E}_x(\omega)$$

Linear Oscillator Model

$$\tilde{P}_x(\omega) = \varepsilon\, \tilde{\chi}_{xx}(\omega) \tilde{E}_x(\omega)$$

$$\vec{\mathrm{P}}(\omega) = \begin{bmatrix} \tilde{P}_x(\omega) \\ \tilde{P}_y(\omega) \\ \tilde{P}_z(\omega) \end{bmatrix} = \tilde{\chi}(\omega)\varepsilon \begin{bmatrix} 3 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} \tilde{E}_x(\omega) \\ \tilde{E}_y(\omega) \\ \tilde{E}_z(\omega) \end{bmatrix}$$

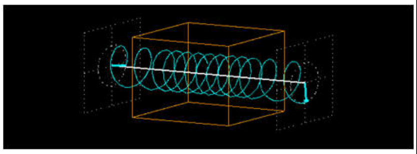
$$\chi(\omega) = -j\frac{1}{4\pi^2}\frac{[N_1 - N_2]\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a}\frac{1}{1 + 2j(\omega - \omega_{0a})/\Delta\omega_a}$$

We have separated the right-hand side into a dimensionless tensor with a trace of magnitude 3, plus a purely scalar susceptibility.

Circularly Polarized Responses

- For a transition that is circularly polarized in the *x*-*y* plane:

$$\vec{\mathrm{P}} = \begin{bmatrix} \tilde{P}_x \\ \tilde{P}_y \\ \tilde{P}_z \end{bmatrix} = \tilde{\chi}(\omega)\,\varepsilon\frac{3}{2}\begin{bmatrix} 1 & \mp j & 0 \\ \pm j & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix}\begin{bmatrix} \tilde{E}_x \\ \tilde{E}_y \\ \tilde{E}_z \end{bmatrix}$$



- If the applied signal is linearly polarized in the *x*-direction:

$$\tilde{E}_x = \tilde{E}_0 \text{ and } \tilde{E}_y = \tilde{E}_z = 0$$

- Then the induced polarization components:

$$\tilde{P}_x = (3\tilde{\chi}\varepsilon/2)\tilde{E}_0 \text{ and } \tilde{P}_y = \mp j(3\tilde{\chi}\varepsilon/)\tilde{E}_0$$

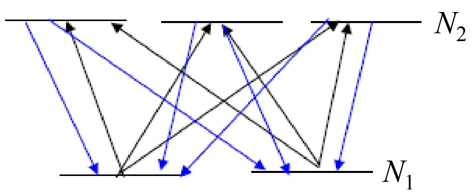
3*

$$\chi(\omega) = -j\frac{3^*}{4\pi^2}\frac{[N_1 - N_2]\lambda^3\lambda_{\text{rad}}}{\Delta\omega_a}\frac{1}{1+2j(\omega-\omega_a)/\Delta\omega_a}$$

- 3* = 3 for fully aligned atoms plus optimally polarized fields
- 3* = 1 either for randomly aligned atoms with arbitrarily polarized fields, or for randomly polarized fields with any atomic alignment
- 3* = 0 for fully aligned atoms and anti-optimum fields
- $0 \leq 3^* \leq 3$ for any intermediate case

15

Degeneracy



- The sub-levels at the same energy are called degenerate.
- In the absence of any magnetic field, all of the m_l levels in the 2p state, i.e., $m_l = 0, \pm 1$ levels have exactly the same energy.
- Remember that we need to add rates. Let the rate from an individual sub-level be $\gamma_{2m\rightarrow 1n}$, where m and n denote the sub-level indices for levels 2 and 1, respectively.

16

Number of Sub-Levels

- Suppose that we have g_2 sub-levels in the upper level (2) and g_1 sub-levels in the lower level (1).
- Usually the number of atoms per unit volume in each sub-level is just N/g.
- Thus we can write the susceptibility for one of the transitions as

$$\overline{\overline{\chi}}_{1n,2m}(\omega) = \tilde{g}(\omega)\gamma_{\text{rad},2m\to 1n}\left(\frac{N_1}{g_1} - \frac{N_2}{g_2}\right)\overline{\overline{T}}_{1n,2m}$$

$$\tilde{g}(\omega) = -j\frac{1}{4\pi^2}\frac{\lambda^3}{\Delta\omega_a}\frac{1}{1+2j\left(\omega-\omega_a\right)/\Delta\omega_a}$$

17

Total Susceptibility

- Here we can now sum up all of the transitions and use the following expressions to define an average T tensor.

$$\overline{\overline{\chi}}_{\text{tot}} = \sum_{n=1}^{g1}\sum_{m=1}^{g_2}\overline{\overline{\chi}}_{1n,2m}(\omega) = \tilde{g}(\omega)\left[\sum_{n=1}^{g_1}\sum_{m=1}^{g_2}\gamma_{\text{rad},2m\to 1n}\left(\frac{N_1}{g_1} - \frac{N_2}{g_2}\right)\right]\overline{\overline{T}}_{\text{av}}$$

- Now we define an average decay rate. To do this let's look at the decay from level 2.

$$\frac{dN_2}{dt} = -\sum_{n=1}^{g_1}\sum_{m=1}^{g_2}\gamma_{\text{rad},2m\to 1n}\left(\frac{N_2}{g_2}\right) = -\left(\frac{N_2}{g_2}\right)\sum_{n=1}^{g_1}\sum_{m=1}^{g_2}\gamma_{\text{rad},2m\to 1n}$$

$$= -N_2\gamma_{\text{rad},2\to 1} \Rightarrow \gamma_{\text{rad},2\to 1} = \frac{1}{g_2}\sum_{n=1}^{g_1}\sum_{m=1}^{g_2}\gamma_{\text{rad},2m\to 1n}$$

18

Correction to ΔN

$$\overline{\overline{\chi}}_{\text{tot}} = \tilde{g}(\omega)\gamma_{\text{rad},2\to1}\left(\frac{g_2 N_1}{g_1} - N_2\right)\overline{\overline{T}}_{\text{av}}$$

$$\chi_{\text{tot}}(\omega) = -j\frac{3^*}{4\pi^2}\frac{\left[\frac{g_2 N_1}{g_1} - N_2\right]\lambda^3\gamma_{\text{rad},2\to1}}{\Delta\omega_a}\frac{1}{1+2j(\omega-\omega_a)/\Delta\omega_a}$$

19

ATOMIC RATE EQUATIONS

20

Power Transfer

- The amount of work dU done by a force f_x acting on the electron, when the electron moves through a distance dx is

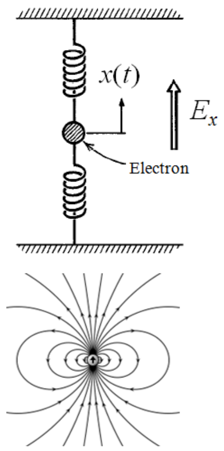
$$dU = f_x\, dx = -eE_x\, dx$$

- Instantaneous rate:

$$\frac{dU(t)}{dt} = -eE_x(t)\frac{dx(t)}{dt} = E_x(t)\frac{d\mu_x(t)}{dt}$$

- Average power flow per unit volume:

$$\frac{dU_a(t)}{dt} = V^{-1}E_x(t)\sum_{i=1}^{NV}\frac{d\mu_{xi}(t)}{dt} = E_x(t)\frac{dp_x(t)}{dt}$$



21

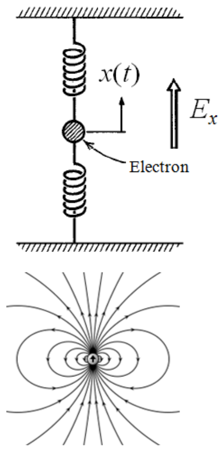
Time-Averaged Power Flow

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

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

$$\tilde{P}(\omega) = \tilde{\chi}(\omega)\varepsilon\tilde{E}(\omega) = \left[\chi'(\omega) + j\chi''(\omega)\right]\varepsilon\tilde{E}(\omega)$$

$$\frac{dU_a(t)}{dt} = E_x(t)\frac{dp_x(t)}{dt} = \frac{1}{4}\left[\tilde{E}(\omega)e^{j\omega t} + \tilde{E}^*(\omega)e^{-j\omega t}\right] \times \left[\tilde{P}(\omega)e^{j\omega t} + \tilde{P}^*(\omega)e^{-j\omega t}\right]$$



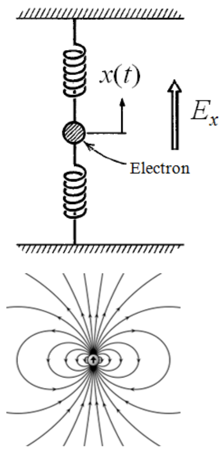
22

Time-Averaged Power Flow

$$\frac{dU_a(t)}{dt} = E_x(t)\frac{dp_x(t)}{dt}$$

$$= \frac{1}{4}\left[\tilde{E}e^{j\omega t} + \tilde{E}^* e^{-j\omega t}\right]\left[\tilde{P}e^{j\omega t} + \tilde{P}^* e^{-j\omega t}\right]$$

$$= \frac{j\omega}{4}\left[\tilde{E}\tilde{P}e^{j2\omega t} - \tilde{E}\tilde{P}^* + \tilde{E}^*\tilde{P} - \tilde{E}^*\tilde{P}^* e^{-j2\omega t}\right]$$



By dropping $e^{\pm j2\omega t}$ terms

$$\frac{dU_a(t)}{dt} = \frac{j\omega}{4}\left[\tilde{E}^*\tilde{P} - \tilde{E}\tilde{P}^*\right]$$

$$= \frac{j\omega}{4}\left[\tilde{E}^*\left(\chi' + j\chi''\right)\varepsilon\tilde{E} - \tilde{E}\left(\chi' - j\chi''\right)\varepsilon\tilde{E}^*\right]$$

$$= -\frac{1}{2}\omega\chi''(\omega)\varepsilon|\tilde{E}|^2$$

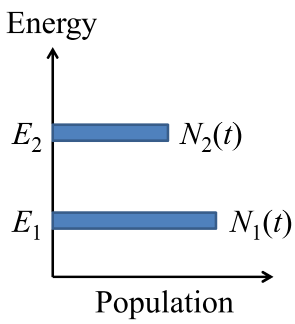
23

Real Quantum Atoms

$$\frac{dU_a(t)}{dt} = -\frac{1}{2}\omega\chi''(\omega)\varepsilon|\tilde{E}|^2$$

$$\tilde{\chi}''(\omega) = -\frac{3^*}{4\pi^2}\frac{\Delta N\lambda^3\gamma_{\text{rad}}}{\Delta\omega_a}\frac{1}{1+\left[2(\omega-\omega_a)/\Delta\omega_a\right]^2}$$

$$\frac{dU_a}{dt} = \left[\frac{3^*}{8\pi^2}\frac{\Delta N\gamma_{\text{rad}}}{\Delta\omega_a}\frac{\omega\varepsilon|\tilde{E}|^2\lambda^3}{1+\left[2(\omega-\omega_a)/\Delta\omega_a\right]^2}\right]$$



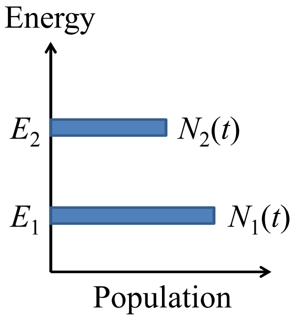
The energy absorption is directly proportional to ΔN.

24

Energy Storage

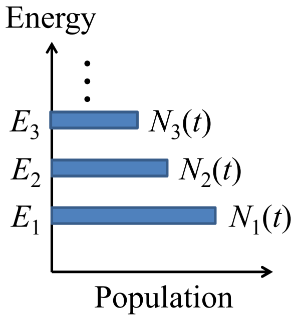
- In a two-level system

$$U_a(t) = N_1(t)E_1 + N_2(t)E_2$$



- In a multilevel system

$$U_a(t) = \sum_{j=1}^{M} N_j(t)E_j$$



Level population N_j changes with time.

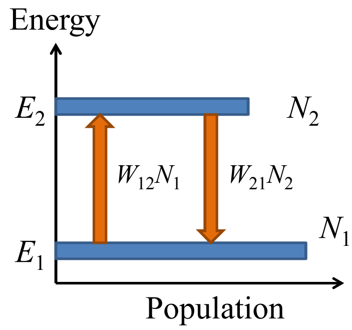
Stimulated Transition Probabilities

$$\frac{dU_a}{dt} = W_{12}N_1\hbar\omega_a - W_{21}N_2\hbar\omega_a$$

$$W_{12} = W_{21}$$

$$= \frac{3^*}{8\pi^2}\frac{\gamma_{\text{rad}}}{\hbar\Delta\omega_a}\frac{\varepsilon|\tilde{E}|^2\lambda^3}{1+\left[2(\omega-\omega_a)/\Delta\omega_a\right]^2}$$

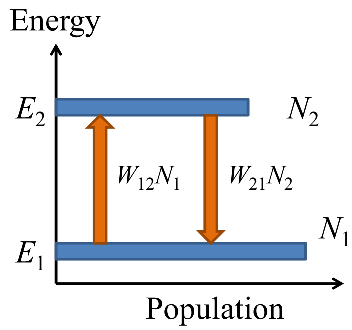
$$W_{12}\hbar\omega_a = W_{21}\hbar\omega_a$$



The energy flow from the signal to the atoms seem to be produced by two flows, i.e., upward stimulated transitions and downward stimulated-transitions of atoms.

Net Flow Rate

$$\left.\frac{dN_2}{dt}\right|_{\text{stim}} = -\left.\frac{dN_1}{dt}\right|_{\text{stim}} = W_{12}N_1 - W_{21}N_2$$



The applied signal gives an atom in the upper level and an atom in the lower level equal probabilities of transitions.

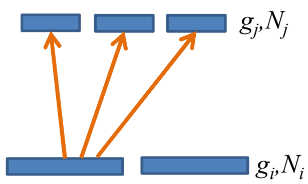
27

Degeneracy

$$\tilde{\chi}''_{ij}(\omega) = -\frac{3^*}{4\pi^2}\frac{\gamma_{\text{rad},ji}\lambda_{ij}^3}{\Delta\omega_a}\frac{\left[(g_j/g_i)N_i - N_j\right]}{1+\left[2\left(\omega-\omega_{ji}\right)/\Delta\omega_{a,ij}\right]^2}$$

Power transfer from signal to atom

$$\frac{dU_a}{dt} = \left[\frac{3^*}{8\pi^2}\frac{\gamma_{\text{rad},ji}}{\Delta\omega_a}\frac{\omega\varepsilon|\tilde{E}_{ij}|^2\lambda_{ij}^3}{1+\left[2\left(\omega-\omega_a\right)/\Delta\omega_a\right]^2}\right] \times\left(\frac{g_j}{g_i}N_i - N_j\right) = W_{ij}N_i\hbar\omega_{ji} - W_{ji}N_j\hbar\omega_{ji}$$



$$W_{ji} = \frac{g_i}{g_j}W_{ij} = \frac{3^*}{8\pi^2}\frac{\gamma_{\text{rad},ji}}{\hbar\Delta\omega_{a,ij}}\frac{\varepsilon|\tilde{E}_{ij}|^2\lambda_{ij}^3}{1+\left[2\left(\omega-\omega_{ji}\right)/\Delta\omega_{a,ij}\right]^2}$$

28