The origin and limitations of the atomic rate equations

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Linear χ

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Chap. 2 resonant-dipole equation (RDE):

$$\frac{d^2P}{dt^2} + \Delta\omega_a \frac{dP}{dt} + \omega_a^2 P = \frac{3^* \omega_a \varepsilon \lambda^3 \gamma_{rad}}{4\pi^2} \Delta N(t) E(t) = K \Delta N(t) E(t)$$

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Was solved in the linear case:

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

$$P(t) = \text{Re}\left\{P_x e^{j\omega t}\right\} = \frac{1}{2}\left(P_x e^{j\omega t} + P_x^* e^{-j\omega t}\right)$$

and using a constant $\Delta N(t) = \Delta N$.

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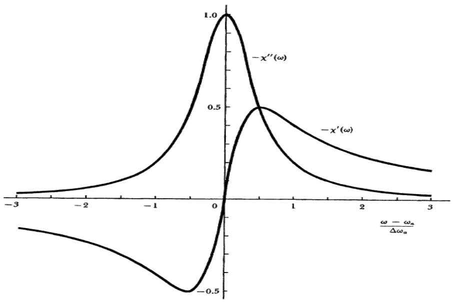
and using a constant $\Delta N(t) = \Delta N$.

This gives:

$$\varepsilon\chi = K \frac{\Delta N}{\omega_a^2 - \omega^2 + j\omega\Delta\omega_a} \rightarrow \text{Lorentzian lineshape}$$



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However, solving the RDE at resonance by applying $E(t) = E_1 \sin\omega_a t$, keeping ΔN constant with $P(0) = P'(0) = 0$ gives:

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However, solving the RDE at resonance by applying $E(t) = E_1 \sin\omega_a t$, keeping ΔN constant with $P(0) = P'(0) = 0$ gives:

let $P = P_h + P_{ih}$, **RHS**(P_h)**=0** **ansatz:** $P_h = Ae^{rt}$, **RHS**(P_{ih})=$Ce^{j\omega_a t}$ **use** $u = ze^{j\omega_a t}$ **and** $\mathrm{Im}\left\{e^{j\omega_a t}\right\}$ =$\sin\omega_a t$

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$$P(t) = -\frac{K\Delta N E_1}{\omega_a \Delta\omega_a}\left[\cos(\omega_a t) - e^{-\frac{\Delta\omega_a}{2}t}\cos\left(\sqrt{\omega_a^2 - \frac{\Delta\omega_a^2}{4}}\,t\right)\right]$$

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$\Delta\omega_a = \gamma + \frac{2}{T_2}$, if $\Delta\omega_a \gg \gamma$ and $\frac{\Delta\omega_a^2}{4} \ll \omega_a^2$, then

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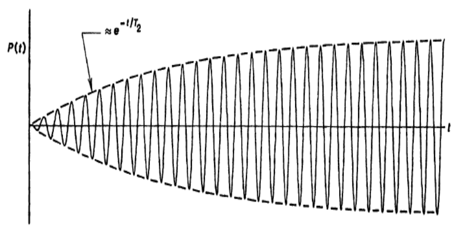
$\Delta\omega_a = \gamma + \frac{2}{T_2}$, if $\Delta\omega_a \gg \gamma$ and $\frac{\Delta\omega_a^2}{4} \ll \omega_a^2$, then

$$P(t) \approx -\frac{K\Delta N E_1}{\omega_a \Delta\omega_a}\left[1 - e^{-\frac{t}{T_2}}\right]\cos(\omega_a t)$$



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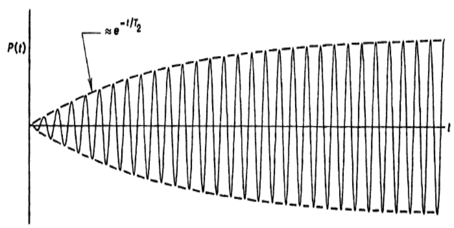

Conclusion:
If $\Delta N(t)$ changes slowly compared to T_2, it can be treated as constant in RDE.

2-level atomic rate equations

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Classical definition of work:

$$dU = \mathbf{F}\cdot d\mathbf{r} = -q\mathbf{E}\cdot d\mathbf{r} \Rightarrow \frac{dU}{dt} = \mathbf{E}\frac{d}{dt}(-q\mathbf{r}) = [\mu = -q\mathbf{r}] = \mathbf{E}\frac{d\mu}{dt}$$

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Average over volume V containing N dipoles:
$\frac{dU_a}{dt} = \mathbf{E}\frac{d}{dt}\left(\frac{1}{V}\sum_{i=1}^{N}\mu_i\right) = \mathbf{E}\frac{d\mathbf{P}}{dt}$

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$$\frac{dU_a}{dt} = \frac{j\omega}{4}\left(\mathbf{E}_1^*\mathbf{P}_1 - \mathbf{E}_1\mathbf{P}_1^*\right) + \frac{j\omega}{4}\left(\mathbf{E}_1\mathbf{P}_1e^{2j\omega t} - \mathbf{E}_1^*\mathbf{P}_1^*e^{-2j\omega t}\right)$$

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At low powers and averaging over a few optical cycles gives:

$$\left.\frac{dU_a}{dt}\right|_{\mathrm{av}} = \frac{j\omega}{4}\left(\mathbf{E}_1^*\mathbf{P}_1 - \mathbf{E}_1\mathbf{P}_1^*\right)$$

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Assuming a linear χ, i.e. $\mathbf{P}_1(\omega) = \varepsilon\chi(\omega)\mathbf{E}_1(\omega)$, gives:

$$\left.\frac{dU_a}{dt}\right|_{\mathrm{av}} = \frac{j\omega\varepsilon}{4}\left(\mathbf{E}_1^*\chi\mathbf{E}_1 - \mathbf{E}_1\chi^*\mathbf{E}_1^*\right)$$

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Let $\mathbf{E}_1\chi^*\mathbf{E}_1^* = \begin{pmatrix} x & y & z \end{pmatrix} \begin{pmatrix} a^* & b^* & c^* \\ d^* & e^* & f^* \\ g^* & h^* & i^* \end{pmatrix} \begin{pmatrix} x^* \\ y^* \\ z^* \end{pmatrix}$

and $\mathbf{E}_1^*\chi^\dagger\mathbf{E}_1 = \begin{pmatrix} x^* & y^* & z^* \end{pmatrix} \begin{pmatrix} a^* & d^* & g^* \\ b^* & e^* & h^* \\ c^* & f^* & i^* \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix}$

$\Rightarrow \mathbf{E}_1\chi^*\mathbf{E}_1^* = \mathbf{E}_1^*\chi^\dagger\mathbf{E}_1$

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Re-express the RHS:

$$\left.\frac{dU_a}{dt}\right|_{\text{av}} = -\frac{j\omega\varepsilon}{4}\mathbf{E}_1^*\left(\chi^\dagger - \chi\right)\mathbf{E}_1 = \left[\chi_{\text{ah}} = \frac{j}{2}\left(\chi^\dagger - \chi\right)\right]$$
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If χ is isotropic (or at least diagonal), then

$\chi^\dagger = \chi^* \Rightarrow \mathbf{E}_1^*\chi_{\text{ah}}\mathbf{E}_1 = -2j\chi''|\mathbf{E}_1|^2$ which gives:

$$\left.\frac{dU_a}{dt}\right|_{\text{av}} = -\frac{\omega\varepsilon}{2}\chi''(\omega)|\mathbf{E}_1(\omega)|^2$$

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This is the average power transfer per unit volume from a sinusoidal field to the atoms in an isotropic and linear medium.

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Using the results in chap. 2, the power transfer equation can be expressed as:

$$\left.\frac{dU_a}{dt}\right|_{\text{av}} = -\frac{\omega\varepsilon}{2}\left[-\frac{3^*\lambda^3\gamma_{\text{rad}}}{4\pi^2\Delta\omega_a}\frac{1}{1+\left(\frac{2(\omega-\omega_a)}{\Delta\omega_a}\right)^2}(N_1-N_2)\right]|\mathbf{E}_1(\omega)|^2$$

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Defining the stimulated upward and downward transition probabilities as: $W_{12} = W_{21} = \frac{3^*\lambda^3\gamma_{rad}}{8\pi^2\Delta\omega_a\hbar}\frac{\varepsilon|\mathbf{E}_1(\omega)|^2}{1+\left(\frac{2(\omega-\omega_a)}{\Delta\omega_a}\right)^2}$

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The energy will be stored in the upper level, so:

$$\frac{dN_2}{dt} = -\frac{dN_1}{dt} = \left.\frac{d}{dt}\left(\frac{1}{\hbar\omega}U_a\right)\right|_{av} = W_{12}N_1 - W_{21}N_2$$

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In the case of degeneracy and $E_i < E_j$, define: $\Delta N = \frac{g_j}{g_i}N_i - N_j$

and change $\left.\begin{array}{c}\gamma_{\text{rad}} \rightarrow \gamma_{\text{rad}\,ij} \\ \lambda \rightarrow \lambda_{ij} \\ \Delta\omega \rightarrow \Delta\omega_{ij} \\ E(\omega) \rightarrow E(\omega_{ij})\end{array}\right\} W_{12} \rightarrow W_{ij}$

Blackbody radiation

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Any volume of space in thermal equilibrium with its surroundings contains blackbody radiation (BBR), if the volume is $\gg \lambda$ the magnitude is given by:

$$d|E_{\mathrm{BBR}}(\omega)|^2 = \frac{16\pi\hbar d\omega}{\varepsilon\lambda^3\left(e^{\frac{\hbar\omega}{kT}}-1\right)}$$

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This field will induce stimulated transitions with transition probabilities given by:

$$W_{12,\mathrm{BBR}} = \int dW_{12,\mathrm{BBR}} = \int_{-\infty}^{\infty} \frac{3^*\lambda^3\gamma_{\mathrm{rad}}}{8\pi^2\Delta\omega_a\hbar}\frac{\varepsilon}{1+\left(\frac{2(\omega-\omega_a)}{\Delta\omega_a}\right)^2}\frac{16\pi\hbar}{\varepsilon\lambda^3\left(e^{\frac{\hbar\omega}{kT}}-1\right)}d\omega$$

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The blackbody spectrum is much broader than the atomic linewidth, $\Delta\omega$, so it can be approximated by its value at the resonance frequency, ω_a, giving:

$$W_{12} = \frac{\gamma_{\text{rad}}}{e^{\frac{\hbar\omega}{kT}}-1}\int_{-\infty}^{\infty}\frac{2}{\pi\Delta\omega_a}\frac{1}{1+\left(\frac{2(\omega-\omega_a)}{\Delta\omega_a}\right)^2}d\omega = \frac{\gamma_{\text{rad}}}{e^{\frac{\hbar\omega}{kT}}-1}$$

Independent of the atomic lineshape!

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Net power absorption?

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$$\frac{\text{energy flow out of the atoms}}{\text{energy flow into the atoms}} = \frac{\left(W_{21,\mathrm{BBR}}+\gamma_{\mathrm{rad}}\right)N_2}{W_{12,\mathrm{BBR}}N_1} =$$

$$\left[W_{12,\mathrm{BBR}} = W_{21,\mathrm{BBR}},\ \gamma_{\mathrm{rad}} + W_{21,\mathrm{BBR}} = W_{21,\mathrm{BBR}}e^{\frac{\hbar\omega}{kT}},\ \frac{N_2}{N_1} = e^{-\frac{\hbar\omega}{kT_a}}\right]$$

$$= e^{\frac{\hbar\omega}{k}\left(\frac{1}{T}-\frac{1}{T_a}\right)} = 1 \text{ if } T = T_a$$

i.e. no net power transfer at thermal equilibrium.

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<u>Detailed balance</u>

Overall thermal equilibrium requires the spontaneous emission rate, given by γ_{rad}, to equal the BBR absorption rate for all transitions and all frequencies in each transition $\Rightarrow$ atomic transitions must have the same lineshapes for spontaneous emission as for stimulated absorption!

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Overall thermal equilibrium requires the spontaneous emission rate, given by γ_{rad}, to equal the BBR absorption rate for all transitions and all frequencies in each transition $\Rightarrow$ atomic transitions must have the same lineshapes for spontaneous emission as for stimulated absorption!

Degeneracy

In the case of degeneracy, the transition rate probabilities are given by:

$$W_{ji,\mathrm{BBR}} = \frac{g_i}{g_j}W_{ij,\mathrm{BBR}} = \frac{\gamma_{\mathrm{rad},ij}}{\mathrm{e}^{\frac{\hbar\omega_{ij}}{kT}}-1}$$

Non-radiative relaxation

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At any finite temperature there will be some motion of atoms, for instance motion of gas particles and lattice vibrations. The collisions and vibrations transfer energy to and from the atoms and can therefore induce stimulated transitions with associated decay rates and transition probabilities:

$$W_{ji,\mathrm{nr}} = \frac{g_i}{g_j} W_{ij,\mathrm{nr}} = \frac{\gamma_{\mathrm{nr},ij}}{e^{\frac{\hbar\omega_{ij}}{kT_{\mathrm{nr}}}} - 1}$$

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This has actually been used to make "acoustic lasers", see for instance:

Phonon Lasing in an Electromechanical Resonator, I. Mahboob, K. Nishiguchi, A. Fujiwara, and H. Yamaguchi, Phys. Rev. Lett. 110, 127202

Phonon Laser Action in a Tunable Two-Level System, Ivan S. Grudinin, Hansuek Lee, O. Painter, and Kerry J. Vahala, Phys. Rev. Lett. 104, 083901

Resulting rate equations

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The transition probabilities resulting from thermal effects can be expressed as:

$$w_{ji} = W_{ji,\mathrm{BBR}} + \gamma_{\mathrm{rad}} + W_{ji,\mathrm{nr}} + \gamma_{ji,\mathrm{nr}}$$
$$w_{ij} = W_{ij,\mathrm{BBR}} + W_{ij,\mathrm{nr}}$$

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$w_{ij} = W_{ij,\mathrm{BBR}} + W_{ij,\mathrm{nr}}$

If $T = T_{\mathrm{nr}}$, the ratio of the transition probabilities is given by:

$\frac{w_{ij}}{w_{ji}} = \frac{g_j}{g_i} e^{-\frac{\hbar\omega_{ji}}{kT}} \Rightarrow w_{ij} < w_{ji}$

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At optical frequencies/wavelengths:

$\frac{\hbar\omega}{k} = [\lambda = 550 \text{ nm}] \approx 26000 \text{ K} \Rightarrow \frac{1}{\mathrm{e}^{\frac{\hbar\omega}{kT}} - 1} = [T = 300 \text{ K}] \approx 0$

which means that the upper level population because of thermal effects will be negligible and:

$w_{ji} \approx \gamma_{ji,\text{rad}} + \gamma_{ji,\text{nr}}$

$w_{ij} \approx 0$

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The origin and limitations of atomic rate equations

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Including the thermal transition probabilities in the 2-level atomic rate equation yields:

$$\frac{dN_1}{dt} = -\frac{dN_2}{dt} = -\left(W_{12} + w_{12}\right) N_1 + \left(W_{21} + w_{21}\right) N_2$$

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Assume no degeneracy and define $N = N_1(t) + N_2(t)$ and $\Delta N(t) = N_1(t) - N_2(t)$, this gives:

$$\frac{d\Delta N}{dt} = -2\left(W_{12} + w_{12}\right) N_1 + 2\left(W_{21} + w_{21}\right) N_2$$

$$= -2W_{12}\Delta N - \left(w_{12} + w_{21}\right)\left(\Delta N - \frac{w_{21} - w_{12}}{w_{12} + w_{21}} N\right)$$

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At thermal equilibrium, $\frac{w_{12}}{w_{21}} = e^{-\frac{\hbar\omega}{kT}} = \frac{N_{20}}{N_{10}}$ which makes $\frac{w_{21} - w_{12}}{w_{12} + w_{21}} N = N_{10} - N_{20} = \Delta N_0$. Using this and defining the relaxation time as $w_{12} + w_{21} = \frac{1}{T_1}$ gives:

$$\frac{d\Delta N}{dt} = -2W_{12}\Delta N - \frac{\Delta N - \Delta N_0}{T_1}$$

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$$\frac{d\Delta N}{dt} = -2W_{12}\Delta N - \frac{\Delta N - \Delta N_0}{T_1}$$

Which can be expressed as:

$$\frac{d\Delta N}{dt} + \frac{\Delta N - \Delta N_0}{T_1} = -2W_{12}\Delta N = -\frac{2}{\hbar\omega}\left.\frac{dU_a}{dt}\right|_{\mathrm{av}}$$

Where the rightmost equal sign shows the rate equations approximation.

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Using the integrating factor, the solution is found to be:

$$\Delta N(t) = \frac{\Delta N_0}{2W_{12}T_1+1} + \left(\Delta N(0) - \frac{\Delta N_0}{2W_{12}T_1+1}\right) e^{-\left(2W_{12}+\frac{1}{T_1}\right)t}$$

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Steady state is obtained as $t \to \infty$, which gives $\Delta N_{ss} = \frac{\Delta N_0}{2W_{12}T_1+1}$.

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Steady state is obtained as $t \to \infty$, which gives $\Delta N_{ss} = \frac{\Delta N_0}{2W_{12}T_1+1}$. As $W_{12} \propto |E|^2$, the population difference will decrease as the signal power increases. This is referred to as homogeneous saturation of the population difference and is what primarily determines the power level lasers will oscillate on, as the gain is proportional to the population difference. It can also be seen that with no applied signal, i.e. $W_{12} = 0$, the population tends to the thermal equilibrium population difference ΔN_0.

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The origin and limitations of atomic rate equations

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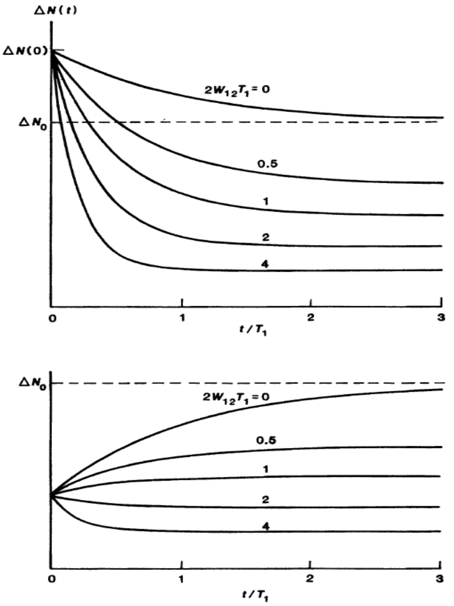
Examples of transient behavior of ΔN



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The requirement that $\Delta N(t)$ changes slowly compared to T_2, or $\Delta\omega_a$ if not simplified, which was assumed when deriving the linear χ, means that:
$2W_{12} + \frac{1}{T_1} \ll \Delta\omega_a = \gamma + \frac{2}{T_2} = [\gamma = \text{decay rate}] = \frac{1}{T_1} + \frac{1}{T_2}$

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Which in turn means that:

- $\frac{1}{T_1} \ll \frac{1}{T_2}$, i.e. the system dephases long before it has relaxed.
- $W_{12} \ll \Delta\omega_a$, i.e. ΔN decays much slower than $P(t)$ reaches steady state.

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For electric dipole transitions $W_{12} = \frac{3^*\lambda^3\gamma_{rad}}{8\pi^2\Delta\omega_a\hbar}\frac{\varepsilon|\mathbf{E}_1(\omega)|^2}{1+\left(\frac{2(\omega-\omega_a)}{\Delta\omega_a}\right)^2}$, which is maximized at resonance, i.e. $\omega = \omega_a$.

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For electric dipole transitions $W_{12} = \frac{3^* \lambda^3 \gamma_{\text{rad}}}{8\pi^2 \Delta\omega_a \hbar} \frac{\varepsilon |\mathbf{E}_1(\omega)|^2}{1 + \left(\frac{2(\omega - \omega_a)}{\Delta\omega_a}\right)^2}$, which is maximized at resonance, i.e. $\omega = \omega_a$. Using this in the latter inequality leads to:
$|E|^2 \ll \frac{\hbar \Delta\omega_a^2}{\gamma_{\text{rad}} \lambda^3 \varepsilon}$
Which also holds for high power lasers that use materials with wide atomic linewidths.

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Saturation occurs when $2W_{12}T_1 \geq 1 \Leftrightarrow W_{12} \geq \frac{1}{2T_1}$.

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In the case of degeneracy, define $\Delta N(t) = \frac{g_2}{g_1}N_1(t) - N_2(t)$ and use $W_{\text{eff}} = \frac{1}{2}(W_{12} + W_{21})$, where $g_1W_{12} = g_2W_{21}$, instead of W_{12}.

Multi-level rate equations

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In real atomic systems there are many energy levels, E_i, with different degeneracies, g_i, and time varying populations, $N_i(t)$. A signal consisting of several frequencies may be near several resonance frequencies and will thus in general induce multiple transitions. If the resonance frequencies differ by a few linewidths, each frequency component will only affect transitions between two levels. In this case, and assuming no interference between the transitions, the RE for each population is given by:
$\frac{dN_t}{dt} = -\sum_{j\neq i}(W_{ij} + w_{ij})\,N_i + \sum_{j\neq i}(W_{ji} + w_{ji})\,N_j$

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$\frac{dN_t}{dt} = -\sum_{j \neq i} (W_{ij} + w_{ij}) N_i + \sum_{j \neq i} (W_{ji} + w_{ji}) N_j$

The relaxations between arbitrary levels are in general very complicated to calculate and are most often guessed at or measured. However, they are always related by: $\frac{w_{ij}}{w_{ji}} = \frac{g_j}{g_i} e^{-\frac{\hbar\omega_{ij}}{kT}}$.

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For k levels, there will be k such equations which can be written in matrix form as:

$$\frac{d}{dt}\begin{pmatrix} N_1 \\ \vdots \\ N_k \end{pmatrix} = \begin{pmatrix} -\sum_{j\neq 1}\left(W_{1j}+w_{1j}\right) & \cdots & \left(W_{k1}+w_{k1}\right) \\ \vdots & \ddots & \vdots \\ \left(W_{1k}+w_{1k}\right) & \cdots & -\sum_{j\neq k}\left(W_{kj}+w_{kj}\right) \end{pmatrix}\begin{pmatrix} N_1 \\ \vdots \\ N_k \end{pmatrix}$$

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If the total number of atoms in all energy levels is constant, there will also be an equation for the conservation of atoms: $\sum_{i=1}^{k} N_i = N$. This results in $k-1$ independent equations and any row in the matrix can be exchanged for a row of ones and the corresponding population to N (use that $\frac{dN}{dt}=0$).

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In steady state, the matrix equation can be solved with gauss elimination. From this it can be found that by increasing the amplitude on one transition but not the others will lead to $\Delta N_{ij,ss} = \frac{\Delta N_{ij,0}}{1+\frac{W_{ij}}{W_{ij,sat}}}$, where the ij-terms on the RHS will depend on the relaxations rates and applied signals on the other transitions. The proof can be found on p. 216-217 in Siegmann.

Simplified large signal analysis

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Starting from the RDE:

$$\frac{d^2P}{dt^2} + \Delta\omega_a \frac{dP}{dt} + \omega_a^2 P = \frac{3^* \omega_a \varepsilon \lambda^3 \gamma_{\text{rad}}}{4\pi^2} \Delta N(t) E(t) = K \Delta N(t) E(t)$$

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And studying it on resonance for $\begin{matrix} E(t) = \mathrm{Re}\left\{E_1(t) e^{j\omega_a t}\right\} \\ P(t) = \mathrm{Re}\left\{-jP_1(t) e^{j\omega_a t}\right\} \end{matrix}$,

the $-j$-factor is because $P(t)$ is -90° out of phase with $E(t)$ at resonance,

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And studying it on resonance for $\begin{array}{c} E(t) = \text{Re}\left\{E_1(t)e^{j\omega_a t}\right\} \\ P(t) = \text{Re}\left\{-jP_1(t)e^{j\omega_a t}\right\} \end{array}$, the $-j$-factor is because $P(t)$ is -90° out of phase with $E(t)$ at resonance, and using that $\Delta\omega_a \ll \omega_a$ and the slowly varying envelope approximation (SVEA), which assumes that $P_1(t)$ doesn't change much in one optical cycle, $\frac{1}{\omega_a}$, such that P_1'' can be omitted leads to:

$$P_1' + \frac{\Delta\omega_a}{2} P_1 = \frac{K\Delta N(t)E_1(t)}{2\omega_a}$$

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$$P_1' + \frac{\Delta\omega_a}{2} P_1 = \frac{K \Delta N(t) E_1(t)}{2\omega_a}$$

From the RE section, we found that:

$$\frac{d\Delta N}{dt} + \frac{\Delta N - \Delta N_0}{T_1} = -\frac{2}{\hbar\omega} \left.\frac{dU_a}{dt}\right|_{\text{av}} = -\frac{2}{\hbar\omega} E \left.\frac{dP}{dt}\right|_{\text{av}}$$

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Starting from the RDE:

$$\frac{d^2P}{dt^2}+\Delta\omega_a\frac{dP}{dt}+\omega_a^2P=\frac{3^*\omega_a\varepsilon\lambda^3\gamma_{\mathrm{rad}}}{4\pi^2}\Delta N(t)E(t)=K\Delta N(t)E(t)$$

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$$\frac{d\Delta N}{dt}+\frac{\Delta N-\Delta N_0}{T_1}=-\frac{2}{\hbar\omega}\left.\frac{dU_a}{dt}\right|_{\mathrm{av}}=-\frac{2}{\hbar\omega}\left.E\frac{dP}{dt}\right|_{\mathrm{av}}$$

Using the same $E(t)$ and $P(t)$ as for the RDE and assuming that $E_1(t)$ and $P_1(t)$ are real yields:

$$\frac{d\Delta N}{dt}+\frac{\Delta N-\Delta N_0}{T_1}=-\frac{1}{\hbar}E_1P_1$$

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Setting $E_1(t) = \begin{cases} E, \ t > 0 \\ 0, \ t < 0 \end{cases}$ and solving the ΔN-equation for P_1 for $t > 0$ yields:

$$P_1 = -\frac{\hbar}{E}\left(\frac{d\Delta N}{dt} + \frac{\Delta N - \Delta N_0}{T_1}\right)$$

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Using this in the modified RDE gives:

$$\left(\frac{d^2}{dt^2} + \left(\frac{\Delta\omega_a}{2} + \frac{1}{T_1}\right)\frac{d}{dt} + \frac{\Delta\omega_a}{2T_1} + \omega_R^2\right)\Delta N(t) = \frac{\Delta\omega_a \Delta N_0}{2T_1}$$

And in a similar manner:

$$\left(\frac{d^2}{dt^2} + \left(\frac{\Delta\omega_a}{2} + \frac{1}{T_1}\right)\frac{d}{dt} + \frac{\Delta\omega_a}{2T_1} + \omega_R^2\right)P(t) = \frac{KE\Delta N_0}{2T_1\omega_a}$$

where the Rabi frequency was defined as $\omega_R = \frac{KE^2}{2\hbar\omega_a}$.

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These decoupled DEs are on the form: $x'' + k_1 x' + k_2 x = f$, where

$$k_1 = \frac{\Delta\omega_a}{2} + \frac{1}{T_1},\ k_2 = \frac{\Delta\omega_a}{2T_1} + \omega_R^2 \text{ and } f = \begin{cases} \frac{\Delta\omega_a \Delta N_0}{2T_1}, & x = \Delta N \\ \frac{KE\Delta N_0}{2T_1\omega_a}, & x = P \end{cases}.$$

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The origin and limitations of atomic rate equations

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Let $x = x_h + x_{ih}$ **where** $\begin{array}{l} x_h'' + k_1 x_h' + k_2 x_h = 0 \\ x_{ih}'' + k_1 x_{ih}' + k_2 x_{ih} = f \end{array}$**. As** f **is a constant,** $x_{ih} = \frac{f}{k_2}$**, use the ansatz** $x_h = Ae^{rt}$**.**

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This gives:

$$x(t) = e^{-\frac{k_1}{2}} \left(A_1 \cosh(\hat{\omega} t) + \frac{A_2}{\sqrt{\frac{\hat{\omega}^2}{|\hat{\omega}^2|}}} \sinh(\hat{\omega} t) \right) + \frac{f}{k_2}$$

where $A_1 = x(0) - \frac{f}{k_2}$, $A_2 = \frac{x'(0) + \frac{k_1}{2}}{\hat{\omega}}$ and $\hat{\omega} = \sqrt{\frac{k_1^2}{4} - k_2}$.

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This gives:

$$x(t) = \mathrm{e}^{-\frac{k_1}{2}}\left(A_1 \cosh(\hat{\omega}t) + \frac{A_2}{\sqrt{\frac{\hat{\omega}^2}{|\hat{\omega}^2|}}}\sinh(\hat{\omega}t)\right) + \frac{f}{k_2}$$

where $A_1 = x(0) - \frac{f}{k_2}$, $A_2 = \frac{x'(0) + \frac{k_1}{2}}{\hat{\omega}}$ and $\hat{\omega} = \sqrt{\frac{k_1^2}{4} - k_2}$.

If $\Delta N(0) = \Delta N_0$ and $\Delta N'(0) = 0$, one finds:

$$\Delta N(t) =$$

$$\frac{\Delta N_0}{1 + \frac{2T_1\omega_R^2}{\Delta\omega_a}}\left(1 + \frac{2T_1\omega_R^2}{\Delta\omega_a}\left(\cosh(\hat{\omega}t) + \frac{1}{\sqrt{\frac{\hat{\omega}^2}{|\hat{\omega}^2|}}}\sinh(\hat{\omega}t)\right)\mathrm{e}^{-\frac{k_1}{2}t}\right)$$

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For a very weak signal, $\omega_R \ll \Delta\omega_a$ and $\frac{1}{T_1} \ll \Delta\omega_a$, one finds that:
$\frac{k_1}{2\hat{\omega}} \approx 1$, $\hat{\omega} - \frac{k_1}{2} \approx -\left(\frac{1}{T_1} + \frac{2\omega_R^2}{\Delta\omega_a}\right)$ and $-\hat{\omega} + \frac{k_1}{2} \approx -\frac{\Delta\omega_a}{2}$.

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This in turn yields:

$$(\cosh(\hat{\omega}t) + \sinh(\hat{\omega}t))\mathrm{e}^{-\frac{k_1}{2}t} \approx \mathrm{e}^{-\left(\frac{1}{T_1} + \frac{2\omega_R^2}{\Delta\omega_a}\right)t} + \mathrm{e}^{-\frac{\Delta\omega_a}{2}t}$$
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$\approx \mathrm{e}^{-\left(\frac{1}{T_1} + \frac{2\omega_R^2}{\Delta\omega_a}\right)t}$, such that:

$$\Delta N(t) = \frac{\Delta N_0}{1+\frac{2T_1\omega_R^2}{\Delta\omega_a}} \left(1 + \frac{2T_1\omega_R^2}{\Delta\omega_a}\mathrm{e}^{-\left(\frac{1}{T_1} + \frac{2\omega_R^2}{\Delta\omega_a}\right)t}\right)$$

$$= \left[\frac{\omega_R^2}{\Delta\omega_a} = \frac{3^*\gamma\text{rad}\varepsilon\lambda^3|E|^2}{8\pi^2\hbar\Delta\omega_a} = W_{12} \text{ @ resonance}\right]$$

$$= \frac{\Delta N_0}{1+2T_1W_{12}} \left(1 + 2T_1W_{12}\mathrm{e}^{-\left(\frac{1}{T_1}+2W_{12}\right)t}\right)$$

which is the RE result.

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If $\omega_R > \frac{\Delta\omega_a}{4} - \frac{1}{2T_1} \Rightarrow \hat{\omega}$ becomes complex and $\begin{matrix}\sinh\\ \cosh\end{matrix} \rightarrow \begin{matrix} j\cdot\sin \\ \cos\end{matrix}$, and $\Delta N(t)$ starts oscillating with a frequency of $\hat{\omega}$, this is referred to as Rabi flopping.

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In the strong signal regime, where $\omega_R \gg \Delta\omega_a$ and $\omega_R \gg \frac{1}{T_1}$, $\hat{\omega} \approx \omega_R$ and $\frac{k_1}{2\hat{\omega}} \approx 0$ so only the cos-term is left. However, the decaying exponential is still left and as $t \rightarrow \infty$ ΔN tends towards the same value as the RE approximation.

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If $\omega_R > \frac{\Delta\omega_a}{4} - \frac{1}{2T_1} \Rightarrow \hat{\omega}$ becomes complex and $\begin{matrix}\sinh\\ \cosh\end{matrix} \rightarrow \begin{matrix} j\cdot\sin\\ \cos\end{matrix}$, and $\Delta N(t)$ starts oscillating with a frequency of $\hat{\omega}$, this is referred to as Rabi flopping.

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The RE condition $W_{12} \ll \Delta\omega_a$ can now be re-expressed as $\omega_R \ll \Delta\omega_a$. This implies that a dephasing or relaxing event is sure to occur and break the Rabi flopping before a cycle is completed.

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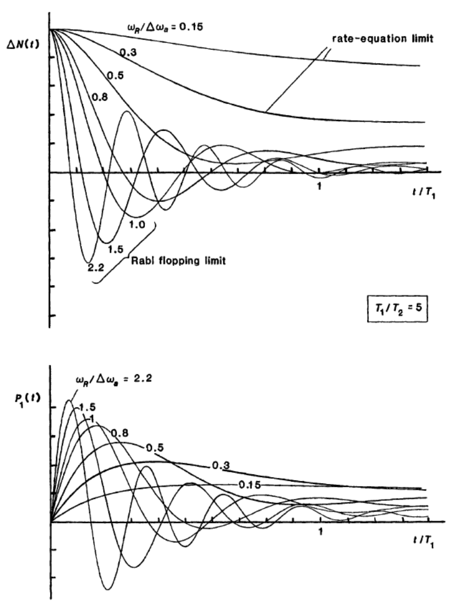
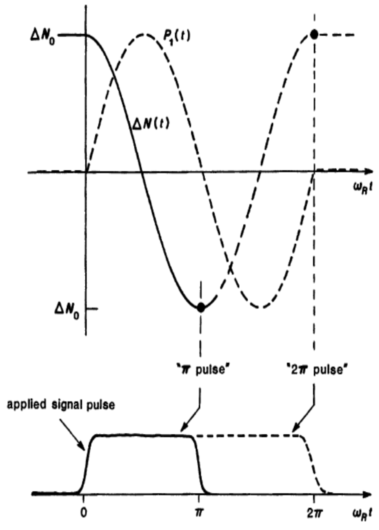
Examples of transient behavior of ΔN





It has experimentally been shown that the inversion can be inverted by applying a pulse of duration T_p such that $\omega_R T_p = \pi$. And also inverted and inverted back if $\omega_R T_p = 2\pi$, which means that the pulse doesn't deliver any energy to the atoms and thus propagates almost attenuation free.

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If an applied signal is strong enough to induce Rabi flopping on some transition $i \rightarrow j$, another signal on for example transition $i \rightarrow k$ will be modulated, by something like the Rabi frequency, due to the moulation of the population from the Rabi flopping.

Problem from the book

Siegmann p.203, Problems for 4.4, task 1

Designed problem

In a two level system, find the symmetrical offset, ω_0, for two sinusoidal signals, i.e. $E = \mathrm{Re}\left\{E_1 e^{j\omega_1 t} + E_2 e^{j\omega_2 t}\right\}$ where $\omega_1 = \omega_a - \omega_0$ and $\omega_2 = \omega_a + \omega_0$, that assures that their combined change in stored energy equals the change in stored energy at resonance. Derive a general expression and apply it to the specific situation when $|E_1|^2 = \frac{1}{4}|E_a|^2$ and $|E_2|^2 = \frac{3}{4}|E_a|^2$ where E_a is the amplitude at resonance.

Hints: Assume low power, $P = \varepsilon\chi E$ and average over a few optical cycles.