QUANTUM ELETRONICS
- BLOEMBERGEN -

Quantum Electronics

Spring 1963, Mo, We, Fr at 11a.m.

Instructor: N. Bloembergen, Pierce Hall 231

Office Hour: After class or by appointment

A thorough background in electromagnetic theory, statistical and quantum mechanics is prerequisite. There is no single required text. The following list of references is recommended.

General

W. Heitler, Quantum Theory of Radiation, Oxford University Press.

J. H. Van Vleck, Theory of Electric and Magnetic Susceptibilities, Oxford University Press.

M. Born and E. Wolf, Principles of Optics, Pergamon Press.

J. H. Griffiths, Theory of Transition Metal Ions, Cambridge University Press.

A. Abragam, Principles of Nuclear Magnetism, Oxford University Press.

H. A. Kramers, Quantum Mechanics.

Introduction to Masers and Lasers

J. R. Singer, Masers, Wiley and Sons.

A. A. Vuylsteke, Elements of Maser Theory, Van Nostrand.

B. A. Langyel, Lasers, Wiley and Sons.

Review of Current Research

Quantum Electronics, edited by C. H. Townes, Columbia University Press, 1960.

Advances in Quantum Electronics, edited by J. R. Singer, Columbia University Press, 1961.

Proceedings IEEE, Quantum Electronics Issue, January 1963.

Supplement on Lasers, Journal of Applied Optics, 1962.

Tentative Outline of Lectures

(No lectures in the week February 11-15, 1963)

Review of the Interaction between Electromagnetic Radiation and Matter.
6 lectures

Quantization of the electromagnetic field
Absorption and stimulated emission
Spontaneous emission, natural line width
Elastic scattering and Raman scattering
Optimum phase definition of waves

Semiclassical Theory of Electric and Magnetic Susceptibilities
6 lectures

Kramers - Heisenberg dispersion formula
Complex susceptibility of ensemble of atoms
Kramers - Kronig causality relation
Local fields and depolarizing fields in dense media
Wave propagation in the medium
Faraday and Kerr effect
Energy levels of paramagnetic ions

Elementary Theory of Masers and Lasers
7 lectures

Phenomenological description of damping
Rate equations and inverted distributions
Negative absorption and dispersion
Circuit representations, negative R, L, and C, traveling wave masers, maser oscillators
Description of lasers, mode selection, coupled rate equations and pulsed operation
Elementary theory of noise
Noise in oscillators

Semiclassical Theory of Non-linear Susceptibilities
4 lectures

Classical models of non-linearity
Non-linear quantum processes, Higher order perturbation theory for two and three level systems
Non-linear complex susceptibilities
Non-linear dielectric tensors

Theory of Parametric Devices
5 lectures

Wave propagation in non-linear media
Parametric amplifiers, up- and-down converters, light modulators
Raman lasers

Exact Solution for Two Level System

4 lectures

Rabi's solution
Ammonia beam maser, hydrogen maser
Coupling between matter and cavity mode

Atomistic Theories of Damping and Relaxation

3 lectures

Spin-lattice relaxation
Inhomogeneous Broadening
Cross-Relaxation

Quantum Noise

4 lectures

Photon statistics in black-body radiation
Coherent oscillators
Coherence functions
Noise in Masers, Parametric Amplifiers and Quantum Counters
Information and Channel Capacity

QUANTUM ELECTRONICS

INSTRUCTOR: BLOEMBERGEN

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Review of the Interaction between EM Radiation and Matter

Classical: Drude, Independent Harmonic oscillator model of electron:

$$m\ddot{x} + m\Gamma\dot{x} + \alpha x = eE(t)$$

or: $$\ddot{x} + \Gamma\dot{x} + \omega_0^2 x = \frac{e}{m} \mathrm{Re}\left[E e^{i\omega t}\right]$$

where $$\omega_0 = \sqrt{\frac{\alpha}{m}}$$

Hence; for the steady state solution:

$$x = \mathrm{Re}\ \frac{(e/m) E e^{-i\omega t}}{-\omega^2 + \omega_0^2 - i\omega\Gamma}$$

This induces a dipole moment: $P(\omega) = e x(\omega)$
This moment has an in-phase and out-phase part. Out-phase = 0 if $\Gamma = 0$.

The instantaneous power absorbed is:

$$eE\dot{x} = eE\cos\omega t\ \mathrm{Re}\ \frac{-i\omega\, e/m\, E e^{-i\omega t}}{-\omega^2 + \omega_0^2 - i\omega\Gamma}$$

$$= \frac{e^2}{m} E^2 \left\{ \frac{\omega^2\Gamma \cos^2\omega t}{(-\omega^2+\omega_0^2)^2 + \omega^2\Gamma^2} - \frac{\omega(-\omega^2+\omega_0^2)\cos\omega t \sin\omega t}{(-\omega^2+\omega_0^2)^2 + \omega^2\Gamma^2} \right\}$$

The time average power absorbed is:

$$\frac{e^2}{2m} E^2 \frac{\omega^2\Gamma}{(-\omega^2+\omega_0^2)^2 + \omega^2\Gamma^2}$$

or; in pure complex notation:

$$\underline{E}(\underline{r},t) = \frac{1}{2}\left[\underline{E}(\underline{r})\, e^{-i\omega t} + \underline{E}^*(\underline{r})\, e^{i\omega t}\right]$$

We assume material of low density, so that applied and local fields are the same. Then:

$$\underline{P}(\underline{r}) = N(\underline{r})\, e\, \underline{\Delta r}$$

where:

$$(\underline{\Delta r})_\ell = \frac{e/m\ E_\ell(\underline{r})}{-\omega^2 + \omega_{0\ell}^2 - i\omega/\tau_\ell}$$

Also: $$\underline{P}(\underline{r},t) = \frac{1}{2}\left[\underline{P}(\underline{r})\, e^{-i\omega t} + \underline{P}^*(\underline{r})\, e^{i\omega t}\right]$$

Then the power absorbed per unit volume is:

$$\underline{E}(\underline{r},t) \cdot \frac{d\,\underline{P}(\underline{r},t)}{dt}$$

The average power is:

$$\frac{\omega}{2\pi} \int_{\text{one cycle}} \underline{E}(\underline{r},t) \cdot \frac{d\underline{P}(\underline{r},t)}{dt}\, dt$$

$$= \frac{\omega}{2\pi}\ \frac{1}{4} \int_0^{\frac{2\pi}{\omega}} \left(\underline{E}(\underline{r})\, e^{-i\omega t} + \underline{E}^*(\underline{r})\, e^{i\omega t}\right)$$

$$\cdot \left(-i\omega\, \underline{P}(\underline{r})\, e^{-i\omega t} + i\omega\, \underline{P}^*(\underline{r})\, e^{i\omega t}\right) dt$$

$$= \omega\, \frac{1}{4}\left(\underline{E}(\underline{r}) \cdot \underline{P}^*(\underline{r}) - \underline{E}^*(\underline{r}) \cdot \underline{P}(\underline{r})\right)$$

$$= \frac{1}{2}\,\omega\, \mathrm{Im}\left(\underline{E}^* \cdot \underline{P}\right) = -\frac{1}{2}\,\omega\, \mathrm{Im}\left(\underline{E} \cdot \underline{P}^*\right)$$

We can write the relation between the polarization and the field as:

$$\underline{P} = (\underline{\underline{\chi}}' + i\,\underline{\underline{\chi}}'') \cdot \underline{E}$$

Then the power absorbed is:

$$\tfrac{1}{2}\,\omega\,\underline{E}^* \cdot \underline{\underline{\chi}}'' \cdot \underline{E} = \tfrac{1}{2}\,\omega\,\chi''\,|E|^2$$

Note that the sign of the absorption depends on the sign convention of $\underline{E}$, but $\underline{\underline{\chi}}''$ changes sign also so absorption is always +. Is there then any classical representation of stimulated emission? Yes, if one suddenly shifts the phase of the field with respect to the polarization by 180°. However, in the steady state the phase is always lagging in the polarization corresponding to positive absorption. Stimulated emission can occur however, only as long as Γ^{-1}, as the field tends to damp the emission. This can be seen by considering the total solution to the equation of motion:

$$x = \mathrm{Re}\left[\frac{e/m\,E\,(e^{-i\omega t} - e^{-i\omega' t})}{-\omega^2 + \omega_0^2 - i\omega\Gamma}\right] + \mathrm{Re}\; x_0\, e^{-i\omega' t}$$

where x_0 is initial displacement and $\omega'^2 = \omega_0^2 - i\omega'\Gamma$. Can see that phase depends on x_0. Problem is to keep stimulated emission in the steady state.

Consider the in-phase part:

stored energy $= \int \underline{E} \cdot d\underline{P} = \tfrac{1}{2}\,\chi' E^2$ with the time average:

$$\tfrac{1}{4}\,\chi'\,\underline{E} \cdot \underline{E}^*$$

NB: $\epsilon' = 1 + 4\pi\chi'$; $\epsilon'' = 4\pi\chi''$

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Recall the development of the classical analogs to stimulated emission and absorption, while Hertz radiation is the analog of spontaneous emission.

We have also introduced the complex susceptibility via:

$$\underline{P} = (\chi' + i\chi'')\underline{E}$$

χ'' + : absorption
χ'' − : stimulated emission

Quantization of the Radiation Field:

Maxwell's Equations for the Vacuum:

$$\underline{E} = -\frac{1}{c}\frac{\partial \underline{A}}{\partial t} - \nabla\phi$$
$$\underline{H} = \text{curl } \underline{A}$$

and: $$\frac{1}{c^2}\frac{\partial^2 \underline{A}}{\partial t^2} - \nabla^2 \underline{A} = \frac{4\pi}{c}\underline{j} \quad ; \quad \underline{j} = \rho \underline{v}$$

$$\frac{1}{c^2}\frac{\partial^2 \phi}{\partial t^2} - \nabla^2 \phi = 4\pi\rho$$

The above are the field form of Maxwell's equations. These are not unique because of the gauge invariance.

$$\nabla^2 \chi - \frac{1}{c^2}\frac{\partial^2 \chi}{\partial t^2} = 0$$

defines: $$\underline{A}' = \underline{A} - \nabla\chi \quad ; \quad \phi' = \phi + \frac{1}{c}\dot{\chi}$$

We will choose the vacuum gauge: $\rho = 0$
$\phi = 0$
$\chi = 0$

which leaves us with:

$$\frac{1}{c^2}\frac{\partial^2 \underline{A}}{\partial t^2} - \nabla^2 \underline{A} = 0$$

In order to solve this equation, we use box normalization, along with BVK boundary conditions: We separate variables and use an eigenfunction expansion:

$$\underline{A}(\underline{r},t) = \sum_{\lambda} q_{\lambda}(t)\ \underline{A}_{\lambda}(\underline{r})$$

giving:

$$\nabla^2 \underline{A}_{\lambda}(\underline{r}) + \frac{\omega_{\lambda}^2}{c^2}\ \underline{A}_{\lambda}(\underline{r}) = 0$$

$$\ddot{q}_{\lambda}(t) + \omega_{\lambda}^2\ q_{\lambda}(t) = 0 \quad \text{(HO equation)}$$

For box normalization, we choose:

$$\underline{A}_{\lambda} = \sqrt{8\pi c^2}\ \begin{matrix}\cos\\ \sin\end{matrix}(\underline{k}_{\lambda}\cdot\underline{r})\ \hat{a}_{\lambda}$$

where $|\underline{k}_{\lambda}| = \frac{\omega_{\lambda}}{c}$ and $\underline{k}_{\lambda}$ is the wave vector. The BC require that:

$$k_{\lambda x} = \frac{2\pi n_x}{L}\ ;\ \text{etc.}$$

$\operatorname{div}\underline{A} = 0$ requires that: $\hat{a}_{\lambda}\cdot\underline{k}_{\lambda} = 0$. λ should be understood as a summation over modes, polarizations, and sine and cosine. We state the orthogonality condition:

$$\frac{1}{L^3}\int \underline{A}_{\lambda}\cdot\underline{A}_{\mu}\,dV = 4\pi c^2\,\delta_{\lambda\mu}$$

The field quantities are now expressed as:

$$\underline{E}(\underline{r},t) = -\frac{1}{c}\sum_{\lambda} \dot{q}_{\lambda}(t)\ \underline{A}_{\lambda}(\underline{r})$$

$$\underline{H}(\underline{r},t) = \sum_{\lambda} q_{\lambda}(t)\ \nabla\times\underline{A}_{\lambda}(\underline{r})$$

To complete the HO statement of q_{λ}, we introduce the canonical variable p_{λ}.

Consider the Hamiltonian for the HO:

$$\mathcal{H}_\lambda = \frac{1}{2}\left(p_\lambda^2 + \omega_\lambda^2 q_\lambda^2\right)$$

$$-\dot{p}_\lambda = \frac{\partial \mathcal{H}_\lambda}{\partial q_\lambda} \quad ; \quad \dot{q}_\lambda = \frac{\partial \mathcal{H}_\lambda}{\partial p_\lambda} = p_\lambda$$

Consider the energy density defined by:

$$U = \frac{1}{8\pi}\int (E^2 + H^2)\, dV$$

Then:

$$\frac{1}{8\pi}\int E^2 dV = \frac{1}{8\pi c^2}\sum_{\lambda\mu} p_\lambda p_\mu \underbrace{\int \underline{A}_\lambda \cdot \underline{A}_\mu\, dV}_{4\pi c^2 \delta_{\lambda\mu}}$$

$$= \frac{1}{2}\sum_\lambda p_\lambda^2$$

This identifies the first half of $\mathcal{H}_\lambda$ with the electric energy density. Now:

$$\frac{1}{8\pi}\int H^2 dV = \frac{1}{8\pi}\sum_{\lambda\mu} q_\lambda q_\mu \underbrace{\int (\nabla\times\underline{A}_\lambda)\cdot(\nabla\times\underline{A}_\mu)\, dV}$$

$$\underbrace{0}_{\text{because of BC}} = \int \underline{A}_\lambda \times (\nabla\times\underline{A}_\mu)\cdot\hat{n}\, dS$$

$$+ \underbrace{\int \underline{A}_\lambda \cdot \nabla\times\nabla\times\underline{A}_\mu\, dV}$$

$$\underbrace{\nabla(\nabla\underline{A}_\mu)}_{0} - \underbrace{\nabla^2\underline{A}_\mu}_{\frac{\omega_\mu^2}{c^2}\underline{A}_\mu}$$

$$= \frac{1}{2}\sum_\lambda \omega_\lambda^2 q_\lambda^2 = \text{second part of } \mathcal{H}_\lambda.$$

Let us now reformulate the situation in complex notation, i.e, we introduce travelling waves instead of standing waves.

We define as a solution:

$$\underline{A}(\underline{r},t) = \sum_{\lambda} \left[q_\lambda(t)\, \underline{A}_\lambda(\underline{r}) + q_\lambda^*(t)\, \underline{A}_\lambda^*(\underline{r}) \right]$$

where:

$$\underline{A}_\lambda(\underline{r}) = \sqrt{4\pi c^2}\, \hat{a}_\lambda\, e^{\imath\, \underline{k}_\lambda \cdot \underline{r}}$$

$$q_\lambda(t) = |q_\lambda|\, e^{-\imath \omega_\lambda t}$$

now here we must have λ going both + and - for traveling waves. This replaces sum over sine, cosine, for standing waves. Orthogonality requires:

$$\int \underline{A}_\lambda \cdot \underline{A}_\mu^*\, dV = 4\pi c^2\, \delta_{\lambda\mu}$$

We now transform to the canonical form by defining:

$$\left.\begin{aligned} Q_\lambda &= q_\lambda + q_\lambda^* \\ P_\lambda &= -\imath\omega\,(q_\lambda - q_\lambda^*) \end{aligned}\right\} \text{ real quantities}$$

We must verify: $\mathcal{H}_\lambda = \frac{1}{2}\left(P_\lambda^2 + \omega_\lambda^2 Q_\lambda^2\right)$ along with the canonical equations. We can also show with the above definitions:

$$\mathcal{H}_\lambda = 2\omega_\lambda^2\, q_\lambda\, q_\lambda^*$$

this will have great advantages when quantizing.

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Recall: $Q_\lambda = q_\lambda + q_\lambda^*$; $P_\lambda = -i\omega_\lambda (q_\lambda - q_\lambda^*)$

$$\mathcal{H}_\lambda = \tfrac{1}{2}\left(P_\lambda^2 + \omega_\lambda^2 Q_\lambda^2\right)$$

$$\dot{P}_\lambda = -\frac{\partial \mathcal{H}_\lambda}{\partial Q_\lambda} \Rightarrow \ddot{Q}_\lambda + \omega_\lambda^2 Q_\lambda = 0$$

$$\dot{Q}_\lambda = \frac{\partial \mathcal{H}_\lambda}{\partial P_\lambda} = P_\lambda$$

For future reference, consider the canonical form of the field vectors $\underline{E}$, $\underline{H}$. Use real quantities:

$$\underline{E}(\underline{r},t) = -\sqrt{4\pi} \sum_\lambda P_\lambda \underline{E}_\lambda(\underline{r})$$

where: $\int \underline{E}_\lambda \cdot \underline{E}_\mu \, dV = \delta_{\lambda\mu}$

$$\underline{H}(\underline{r},t) = \sqrt{4\pi} \sum_\lambda \omega_\lambda q_\lambda \underline{H}_\lambda(\underline{r})$$

so that: $\int \underline{H}_\lambda \cdot \underline{H}_\mu \, dV = \delta_{\lambda\mu}$

We now proceed to quantize the field. Use the Dirac method:

$$[P_\lambda, Q_\mu] = P_\lambda Q_\lambda - Q_\mu P_\lambda = -i\hbar$$

$$[P_\lambda, Q_\mu] = -i\hbar\,\delta_{\lambda\mu}$$

For the HO: $W_\lambda = (n_\lambda + \tfrac{1}{2})\hbar\omega_\lambda$ where we know that the selection rules permit transitions between adjacent states:

$$\int \psi_n^*(\xi)\, Q_\lambda\, \psi_{n+1}(\xi)\, d\xi = Q_{n,n+1} = \left(\frac{\hbar(n+1)}{2\omega_\lambda}\right)^{1/2} e^{-i\omega_\lambda t}$$

$$= Q_{n+1,n}^*$$

From this we deduce:

$$q_{n,n+1} = \left(\frac{\hbar (n+1)}{2\omega}\right)^{1/2} e^{-i\omega t}$$

$$q^*_{n+1,n} = \left(\frac{\hbar (n+1)}{2\omega}\right)^{1/2} e^{i\omega t}$$

$$q_{n+1,n} = 0$$

$$q_\lambda q^*_\mu - q^*_\mu q_\lambda = \frac{\hbar}{2\omega_\lambda}\delta_{\lambda\mu}$$

We can show that in each eigenstate $\langle E^2(x,t)\rangle$ and $\langle H^2(x,t)\rangle$ are completely determined since they are $\frac{1}{2}W_\lambda$. However, $\langle E(x,t)\rangle = 0$ since we really consider $\langle p_\lambda\rangle$. In other words, since we have specified W_λ exactly, we know nothing about the phase and if we average over E over all phases, we get zero. Same with H. One can show that $|E|$ and its phase are conjugate variables and obey the uncertainty principle:

$$\Delta P \, \Delta Q \geq \frac{\hbar}{2} \quad \text{where} \quad \Delta P = \left[\langle P^2\rangle - \langle P\rangle^2\right]^{1/2}, \text{ etc.}$$

Also: $\Delta W_\lambda \, \Delta t \geq \frac{\hbar}{2}$ or: $\Delta \omega_\lambda \, \Delta t \geq 1$

One must interpret this a little differently for the HO. Write:

$$\Delta \frac{W_\lambda}{\hbar \omega_\lambda} \, \Delta \omega_\lambda t \geq \frac{1}{2} \quad \text{or} \quad \Delta n_\lambda \, \Delta \varphi_\lambda \geq \frac{1}{2}$$

We must form a wave packet (linear combination of eigenstates) in order to get a determination of the phase. The proper combination will give <u>optimum phase definition</u>. This is an exercise in Ehrenfest's theorem, that is, we find that combination that closely describes the classical case. This is the problem of the minimum uncertainty wave packet.

For a continuous distribution we would have a Gaussian. But here we have:

$$\psi = \sum_n C_{n_s} |n_s\rangle$$

which is discreate. This suggest we use instead the Poisson distribution. We have for the minimum uncertainty wave packet or optimum definition of amplitude plus phase:

$$\psi(t) = \sum_n C_n e^{-i\omega t(n+1/2)} |n\rangle$$

where $$C_n = \frac{[e^{-\bar{n}}(\bar{n})^n]^{1/2} e^{-in\varphi}}{[n!]^{1/2}}$$

Recall for time independant solution of the HO:

$$u_n(q) = \left(\frac{\alpha}{\pi^{1/2} 2^n n!}\right)^{1/2} H_n(\alpha q) e^{-1/2 \alpha^2 q^2}$$

where:

$$H_n(\alpha q) = (-1)^n e^{(\alpha q)^2} \frac{d^n\{e^{-(\alpha q)^2}\}}{d(\alpha q)^n}$$

$$\alpha = (\hbar^{-1}\omega)^{1/2}$$

We may now check our heuristic results above by considering:

$$\langle p\rangle = \int \psi^* \frac{\hbar}{i} \frac{\partial}{\partial q} \psi \, dq = (2\bar{n}\hbar\omega)^{1/2} \sin(\omega t + \varphi)$$

and $$\langle q\rangle = (2\bar{n})^{1/2} \left(\frac{\hbar}{\omega}\right)^{1/2} \cos(\omega t + \varphi)$$

We must now show that this is the best we can do and also calculate $\langle E^2\rangle$: We find:

$$[\langle p^2\rangle - \langle p\rangle^2][\langle q^2\rangle - \langle q\rangle^2] = \frac{\hbar^2}{4}$$ which is the best we can do, so we have found the best wave packet.

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Continuation of Phone Discussion: Classically, the phase is specified by giving the initial momentum and position:

$$Q = Q_0 \cos\omega t + \frac{P_0}{\omega}\sin\omega t = A\cos(\omega t + \phi)$$
$$P = -Q_0\,\omega \sin\omega t + P_0 \cos\omega t$$

Hence: $\phi = \arctan \dfrac{P_0}{\omega Q_0}$

We now consider a classical statistical distribution in P_0 and Q_0, and consider the fluctuation small. Hence:

$$\Delta\phi = \frac{\omega Q_0 \Delta P_0 - \omega P_0 \Delta Q_0}{\omega_0^2 Q_0^2 + P_0^2}$$

Now find: $\overline{\Delta\phi^2}$. We must assume something for the distribution of P_0 and Q_0 and that these distributions are uncorrelated. Hence:

$$\left|\frac{\Delta Q_0}{Q_0}\right| \ll 1 \; ; \quad \left|\frac{\Delta P_0}{P_0}\right| \ll 1 \; ; \quad \overline{\Delta P_0\,\Delta Q_0} = 0$$

Also assume: $\overline{\Delta P_0^2} = \omega^2 \overline{\Delta Q_0^2}$ which means the magnitudes of the fluctuation are equal, which we will find in the condition of the minimum uncertainty principle. We find:

$$\overline{\Delta Q^2} = (\overline{Q_0^2} - \overline{Q_0}^2)\cos^2\omega t + \frac{1}{\omega^2}(\overline{P_0^2} - \overline{P_0}^2)\sin^2\omega t$$
$$= \overline{Q_0^2} - \overline{Q_0}^2$$

Since $\overline{\Delta Q^2}$ is independent of time, we have a stationary packet. Therefore:

$$\overline{\phi^2} - \overline{\phi}^2 = \frac{\omega^2 Q_0^2\,\overline{\Delta P_0^2} + \omega^2 P_0^2\,\overline{\Delta Q_0^2}}{(\omega^2 Q_0^2 + P_0^2)^2} = \frac{\overline{\Delta P_0^2}}{\omega^2 Q_0^2 + P_0^2}$$

$$= \frac{\overline{\Delta P_0^2}}{2W}$$

Now: $\Delta W = \frac{1}{2}\omega^2 \Delta Q_0^2 + \frac{1}{2}\Delta P_0^2$

$$\overline{W^2} - \overline{W}^2 = \overline{\Delta W^2} + 2W\overline{\Delta W} \approx 2W\overline{\Delta P_0^2} = \hbar^2\omega^2\overline{\Delta n^2}$$

Classically, we do not know how large fluctuations should be but in QM we know: $\overline{\Delta P_0^2} = \frac{\hbar\omega}{2}$ at least, since this leads to:

$$\sqrt{\overline{\Delta P^2}\ \overline{\Delta Q^2}} = \frac{\hbar}{2}$$

Resubstituting, we have:

$$\overline{\Delta n^2} = \frac{2W\ \frac{\hbar\omega}{2}}{\hbar^2\omega^2} = \frac{W}{\hbar\omega} = \bar{n}$$

Hence:

$$\overline{\Delta\phi^2} = \frac{\hbar\omega}{2\cdot 2\bar{n}\hbar\omega} = \frac{1}{4\bar{n}}$$

which gives:

$$\sqrt{\overline{\Delta n^2}\ \overline{\Delta\phi^2}} = \frac{1}{2}$$

We have developed the classical analog of the minimum uncertainty principle.

Now, if in QM we want a state with some phase, we must form a linear combination of pure energy states. To do this, we take the coefficients to be Poisson distributed according to some average number of quanta $\bar{n}$. Recall:

$$\psi_{\substack{\text{min.}\\ \text{unc.}}} = \sum_{n=0}^{\infty} C_n e^{-i\omega t(n+1/2)} |n\rangle$$

where:

$$C_n = \left(\frac{e^{-\bar{n}}(\bar{n})^n}{n!}\right)^{1/2} e^{-in\phi}$$

and:

$$\bar{n} = \sum_{n=0}^{\infty} |C_n|^2 n$$

ϕ here defines the phase.

What we now want is some expectation values at $t=0$. Now note:

$$|n\rangle = \frac{\left(\frac{2\omega}{\hbar}\right)^{n/2} q^n}{\sqrt{n!}} |0\rangle = \frac{a^{+n}}{\sqrt{n!}} |0\rangle$$

can be written in terms of the creation operator. This is the approach used in the paper:

Louisell, Yariv, and Siegman, PR124, 1646 (1961). See Appendices I and II.

Hence we write:

$$\psi_{min} = e^{-\bar{n}/2} \sum_{n=0}^{\infty} \frac{(\bar{n}^{1/2} e^{-i\phi} a^+)^n}{n!} |0\rangle$$

$$= e^{-\bar{n}/2} \exp(\omega a^+) |0\rangle$$

Now we want $Exp\left[(q^* + q)\left(\frac{2\omega}{\hbar}\right)^{1/2}\right] = Exp\left[a^+ + a\right]$:

$$(\psi^*_{min} / a^+ / \psi_{min}) = e^{-\bar{n}} \langle 0| \exp(\omega^* a)\, a^+ \exp(\omega a^+) |0\rangle$$

where $\omega^* \equiv \bar{n}^{1/2} e^{+i\phi}$; $aa^+ - a^+a = 1$; $a = \frac{\partial}{\partial a^+}$

We use the result:

$$e^{ua} f(a^+) = f(a^+ + u)$$

Hence: $(\psi^*_{min} / a^+ / \psi_{min}) = e^{-\bar{n}} \langle 0| (a^+ + \omega^*) \exp\{\omega (a^+ + \omega^*)\} |0\rangle$

We see: $\langle 0| a^+ \exp(\omega a^+ + \omega\omega^*) |0\rangle = 0$

Consider:

$$\langle 0| \omega^* \exp(\omega a^+ + \omega\omega^*) |0\rangle = \omega^* \exp(\omega\omega^*)$$
$$= \omega^* e^{\bar{n}}$$

And:

$$(\psi^*_{min} / a^+ / \psi_{min}) = \omega^* = \bar{n}^{1/2} e^{i\phi}$$

Also:

$$(\psi^*_{min} / a / \psi_{min}) = \omega = \bar{n}^{1/2} e^{-i\phi}$$

Then:

$$\langle Q \rangle = \left(\frac{\hbar}{2\omega}\right)^{1/2} 2\bar{n}^{1/2} \cos\phi$$

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The Interaction Between EM Fields and Matter

$$\mathcal{H} = \mathcal{H}_{rad} + \mathcal{H}_{matter} + \mathcal{H}_{int}$$

We will treat $\mathcal{H}_{int}$ as a small perturbation. Now:

$$\mathcal{H}_{matter} = \sum_k \frac{p_k^2}{2m_k} + \sum_{jk}{}' \frac{e_j e_k}{r_{jk}}$$

We will use the one-particle non-relativistic approximation, that is, we use the Hartree-Fock approximation. Hence:

$$\mathcal{H}_{matter} = \frac{p^2}{2m} + e\phi_k$$

In the presence of a radiation field:

$$\mathcal{H} = \frac{\left(\underline{p} - \frac{e}{c}\underline{A}\right)^2}{2m} + e\phi$$

where the field is described by the vector potential $\underline{A}$. Now:

$$\mathcal{H} = \mathcal{H}_{matter} - \frac{e}{mc}\,\underline{p}\cdot\underline{A} + \frac{e^2}{2mc^2}\,\underline{A}\cdot\underline{A}$$

Recall that when $\operatorname{div}\underline{A} = 0$, $\underline{p}$ and $\underline{A}$ commute. The last two terms comprise $\mathcal{H}_{int}$. $\mathcal{H}_{rad}$ is given by:

$$\mathcal{H}_{rad} = \sum_\lambda \omega_\lambda^2 \left(q_\lambda q_\lambda^* + q_\lambda^* q_\lambda \right)$$

where:

$$\underline{A}(\underline{s},t) = \sum_\lambda q_\lambda \hat{a}_\lambda (4\pi c^2)^{1/2} e^{i\underline{k}_\lambda\cdot\underline{s}} e^{-i\omega_\lambda t} + \sum_\lambda q_\lambda^* \hat{a}_\lambda (4\pi c^2)^{1/2} e^{-i\underline{k}_\lambda\cdot\underline{s}} e^{i\omega_\lambda t}$$

We now investigate the linear term in $\mathcal{H}_{int}$. We see that this can only change the number of quanta in the field by ± 1.

Consider the following matrix element:

$$\mathcal{H}'_{a\, n_\lambda,\ b\, n_\lambda+1} = -\left(\frac{2\pi c^2 \hbar}{\omega_\lambda}\right)^{1/2} \left(\frac{e}{mc}\right) (n_\lambda+1)^{1/2}\, e^{-i\omega_\lambda t}$$

$$\cdot \int \psi_a^* (\mathbf{p}\cdot\hat{a}_\lambda)\, e^{i\mathbf{k}_\lambda\cdot\mathbf{r}}\, \psi_b\, d\tau$$

$$\mathcal{H}'_{a\, n_\lambda+1,\ b\, n_\lambda} = -\left(\frac{e}{mc}\right)\left(\frac{2\pi c^2\hbar}{\omega_a}\right)^{1/2} (n_\lambda+1)^{1/2}\, e^{i\omega_a t}$$

$$\cdot \int \psi_a^* (\mathbf{p}\cdot\hat{a}_\lambda)\, e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}}\, \psi_b\, d\tau$$

Now examine the quadratic term. This term can change the number of quanta by $+2$, 0, or -2. Consider:

$$\mathcal{H}'_{a\, n_\lambda+1\ n_\mu+1,\ b\, n_\lambda\, n_\mu} = \frac{e^2}{2m}(\hat{a}_\lambda\cdot\hat{a}_\mu)\frac{2\pi\hbar}{(\omega_\lambda\omega_\mu)^{1/2}} (n_\lambda+1)^{1/2}(n_\mu+1)^{1/2}$$

$$\cdot \int \psi_a^*\, e^{-i(\mathbf{k}_\lambda+\mathbf{k}_\mu)\cdot\mathbf{r}}\, \psi_b\, d\tau\ e^{i(\omega_\lambda+\omega_\mu)t}$$

This gives only one ME but the others are similar.

Although this is a two photon process, in second order perturbation theory, the linear term gives a two photon process.

Let the matter states be those of the free electron, characterized by the momenta $\mathbf{k}_a$ and $\mathbf{k}_b$. We see that the law of conservation of momentum arises, i.e., $\mathbf{k}_a - \mathbf{k}_\lambda - \mathbf{k}_\mu - \mathbf{k}_b = 0$. This law does not exist for bound electrons because momentum can be taken up by the nucleus also.

Now we may also consider the effects of magnetic fields characterized by the vector potential A_H and the interaction with a molecular magnetic field (Weiss field). Hence:

$$\frac{\left(\mathbf{p} - \frac{e}{c}\mathbf{A}_H - \frac{e}{c}\mathbf{A}\right)^2}{2m} + e\phi$$

$$= \underbrace{\frac{\left(\mathbf{p} - \frac{e}{c}\mathbf{A}_H\right)^2}{2m} + e\phi}_{\mathcal{H}_{\text{matter}}} - \frac{e}{2mc}\left(\mathbf{p} - \frac{e}{c}\mathbf{A}_H\right)\cdot\mathbf{A} + \frac{e^2}{2mc^2}\mathbf{A}\cdot\mathbf{A}$$

In the ME's, this means:

$$\underline{p} \cdot \hat{a}_\lambda \rightarrow \left(\underline{p} - \frac{e}{c}\underline{A}_H\right) \cdot \hat{a}_\lambda$$

Consider:

$$\underline{v} = \dot{\underline{r}} = \frac{1}{i\hbar}\left(\mathcal{H}_{matter}\,\underline{r} - \underline{r}\,\mathcal{H}_{matter}\right)$$

$$= \frac{1}{m}\left(\underline{p} - \frac{e}{c}\underline{A}_H\right)$$

Then the linear interaction term is: $-\frac{e}{c}\,\underline{v}\cdot\underline{A}$
We see that the interaction is most generally specified by the operator $\underline{v}$ rather than the operator $\underline{p}$.

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Time Dependent Perturbation Theory: Motion of a material system under a perturbation which is a random function of the time (damping terms). Consider:

$$i\hbar\,\dot{\psi} = \mathcal{H}(t)\,\psi + \mathcal{H}_0\,\psi$$

with $\mathcal{H}(t)$ the perturbation. The general solution is:

$$\psi = \sum_n a_n(t)\, e^{-i E_n t/\hbar}\, \varphi_n$$

where: $\mathcal{H}_0 \varphi_n = E_n \varphi_n$. Resubstitution gives:

$$i\hbar\,\dot{a}_n = \sum_m \mathcal{H}_{nm}(t)\, a_m(t)\, e^{i\omega_{nm} t}$$

;

$$\omega_{nm} = \frac{E_n - E_m}{\hbar} \; ; \qquad \mathcal{H}_{nm} = \int \varphi_n^*\, \mathcal{H}(t)\, \varphi_m\, dV$$

We solve by successive approximations. Assume at $t=0$ we are in a definite state so that at $t=0$: $a_m(0) = 1$; $a_{n'}(0) = 0$, $n' \neq m$. Then:

$$a_n(t) = \frac{1}{i\hbar} \int_0^t \mathcal{H}_{nm}(t)\, e^{i\omega_{nm} t}\, dt$$

$$P_{nm}(t) = |a_n(t)|^2 = \frac{1}{\hbar^2} \int_0^t \int_0^t \mathcal{H}_{nm}(t')\, \mathcal{H}^*_{nm}(t'')\, e^{i\omega_{nm}(t'-t'')}\, dt'\, dt''$$

We now take a statistical average over an ensemble of perturbations. Make the substitutions $\tau = t' - t''$ and $T = t' + t''$. Then:

$$\bar{P}_{nm}(t) = \frac{1}{2\hbar^2} \int_0^{2t} dT \int d\tau\; G(\tau)\, e^{i\omega_{nm} t}$$

where $G(\tau) = \overline{\mathcal{H}_{nm}(t')\, \mathcal{H}^*_{nm}(t'')}$ and in an even stationary random process, hence $G(\tau) = G^*(-\tau)$.

Consider only times t that are much greater than the correlation time τ_0, that is, $G(\tau) = 0$ for $\tau \gg \tau_c$. Usually τ_c arises from a dependence like $e^{-|\tau|/\tau_c}$.

This means the integration limits on τ can be taken from $+\infty$ to $-\infty$. This leads to an error of τ_c/t. Hence we get a function that is linearly dependent on t as per the usual treatment of time perturbation theory.

We now define the transition rate:

$$w_{nm} = \frac{d\,\overline{P_{nm}(t)}}{dt} \qquad \frac{J(\omega_{nm})}{\hbar^2}$$

where:

$$J(\omega) = \int_{-\infty}^{\infty} G(\tau)\, e^{i\omega\tau}\, d\tau$$

Now, quite often the perturbation takes the form:

$$\mathcal{H}(t) = O_{op}\, F(t)$$

Then:

$$w_{nm} = \frac{|O_{op}|^2}{\hbar^2}\, f(\omega_{nm})$$

where $f(\omega) = \int_{-\infty}^{\infty} \overline{F(t)\, F(t-\tau)}\, e^{i\omega\tau}\, d\tau$

In the usual case or treatment, f is the density of final states.

We now remove the restriction on the definiteness of the $t=0$ state. We do this by introducing the density matrix.

$$\rho_{nn'} = a_n a_{n'}^* \quad ; \quad i\hbar\,\dot{a}_n = \sum_m (\mathcal{H}_{nm} + \mathcal{H}^0_{nm})\, a_m$$

Then:

$$i\hbar\,\dot{a}_n a_{n'}^* = \sum_m \mathcal{H}^{tot}_{nm}\, a_m a_{n'}^*$$

$$i\hbar\,\dot{a}_{n'}^* a_n = -\sum_m \mathcal{H}_{n'm}\, a_m^* a_n$$

from which we get:

$$i\hbar\,\dot{\rho}_{nn'} = [\mathcal{H}^{tot}, \rho]_{nn'}$$

Again consider: $\mathcal{H}^{tot} = \mathcal{H}_0 + \mathcal{H}(t)$. Then:

$$i\hbar\,\dot{\rho} = \left[\mathcal{H}_0 + \mathcal{H}(t),\, \rho\right]$$

Now transform to the interaction representation by:

$$\rho' = e^{\frac{i}{\hbar}\mathcal{H}_0 t}\,\rho\, e^{-\frac{i}{\hbar}\mathcal{H}_0 t}$$

$$\mathcal{H}'(t) = e^{\frac{i}{\hbar}\mathcal{H}_0 t}\,\mathcal{H}(t)\, e^{-\frac{i}{\hbar}\mathcal{H}_0 t}$$

Then:

$$i\hbar\,\dot{\rho}' = \left[\mathcal{H}'(t),\, \rho'\right]$$

We now integrate:

$$i\hbar\,\rho'(t) = i\hbar\,\rho'(0) + \int_0^t \left[\mathcal{H}'(t'),\, \rho'(0)\right] dt'$$

$$+ \frac{1}{i\hbar}\int_0^t \int_0^{t'} \left[\mathcal{H}'(t'),\, \left[\mathcal{H}'(t''),\, \rho'(0)\right]\right] dt'\,dt'' + \cdots$$

We now take an ensemble average over the time dependent density matrix. For example, the initial condition may be thermal equilibrium. Note: there may be a correlation between $\rho'(0)$ and $\mathcal{H}'(t')$. However, for $t' \gg \tau_c$, the correlation vanishes. This allows us in the integration to treat $\rho'(0)$ and $\mathcal{H}'(t)$ to be uncorrelated and we can take separate averages. However, $\overline{\mathcal{H}'(t')} = 0$ so that the second term vanishes.

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It is to be emphasized that the density matrix method need not be used but that the same results could be obtained in terms of the "a" coefficients. Recall:

$$i\hbar\,\dot{a}_n = \sum_m H_{nm}(t)\, e^{i\omega_{nm}t}\, a_m$$

If we take $a_m(0) = 1$, we found:

$$a_n = \frac{1}{i\hbar}\int_0^t H_{nm}(t')\, e^{i\omega_{nm}t'}\, dt'$$

We look at the initial state:

$$a_m = \frac{1}{i\hbar}\sum_n \int_0^t H_{mn}(t')\, a_n(t')\, e^{i\omega_{mn}t'}\, dt'$$

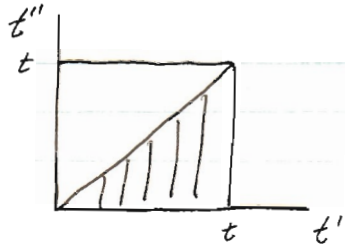
We have excluded time dependent diagonal elements in the second approximation:

$$a_m(t) = -\frac{1}{\hbar^2}\sum_n \int_0^t\int_0^{t'} H_{mn}(t')\, H_{nm}(t'')\, e^{i\omega_{mn}(t'-t'')}\, dt'\,dt''$$

Note that this is similar to:

$$a_n(t)\, a_n^*(t) = -\int_0^t\int_0^t H_{nm}(t)\, H_{nm}^*(t'')\, e^{-i\omega_{nm}(t'-t'')}\,\frac{dt'\,dt''}{\hbar^2}$$
$$= w_{nm}\, t$$

except for the upper limits on the second $\int$. This amounts to the difference in integrating over a square or a triangle:



Hence:

$$a_m(t) = -\frac{1}{2}\, a_m(0)\sum_n w_{nm}\, t + a_m(0)$$

or:

$$\dot{a}_m = -a_m\,\frac{1}{2}\sum_n w_{nm}$$

The solution is:

$$a_m = a_m(0)\, e^{-\frac{1}{2}\Gamma_{mm} t} \quad ; \quad \Gamma_{mm} = \sum_n W_{nm}$$

This was first expounded by Weisskopf and Wigner 1930 and reviewed by Breit, Rev. Mod. Phys. 5, 91 (1933). Since we now know the behaviour of the a's with time, we also know the behaviour of the density matrix with time, that is:

$$\rho_{nm}(t) = a_n(t)\, a_m^*(t) \propto e^{-\frac{1}{2}(\Gamma_{mm}+\Gamma_{nn})t}$$

and $$\dot{\rho}_{nm}(t) = \rho_{nm}(0)\, e^{-\frac{1}{2}(\Gamma_{mm}+\Gamma_{nn})t}$$

when we have a random perturbation with no diagonal terms.

We now consider:

$$\dot{\rho} = \frac{1}{i\hbar}[\mathcal{H}, \rho] \quad ; \quad \mathcal{H} = \mathcal{H}_0 + \mathcal{H}(t)$$

which we can write:

$$\dot{\rho} = \frac{1}{i\hbar}[\mathcal{H}_0, \rho] - \frac{1}{2}(\Gamma\rho + \rho\Gamma)$$

This is for a single atomic system. Since the equation is linear, we can take the ensemble average over both sides. Γ is the damping factor arising from the ensemble average over the random perturbing Hamiltonian:

$$\dot{\bar{\rho}} = \frac{1}{i\hbar}[\mathcal{H}_0, \bar{\rho}] - \frac{1}{2}(\Gamma\bar{\rho} + \bar{\rho}\Gamma)$$

We now transform to primed system as in last time:

$$i\hbar\,\dot{\rho}' = [\mathcal{H}', \rho']$$

$$\rho' = \exp\left(i\frac{\mathcal{H}_0}{\hbar}t\right)\rho\exp\left(-i\frac{\mathcal{H}_0}{\hbar}t\right)$$

$$\mathcal{H}' = \exp\left(i\frac{\mathcal{H}_0}{\hbar}t\right)\mathcal{H}(t)\exp\left(-i\frac{\mathcal{H}_0}{\hbar}t\right)$$

$$\rho'(t) = \rho'(0) + \frac{1}{i\hbar}\int_0^t [\mathcal{H}', \rho'(0)]\,dt' + \frac{1}{-\hbar^2}\int_0^t dt' \int_0^{t'} dt''$$
$$\cdot [\mathcal{H}'(t'), [\mathcal{H}'(t''), \rho'(0)]]$$

We average over the initial conditions, considering times longer than the correlation time.

$$\bar{\rho}'(t) = \bar{\rho}'(0) - \frac{t}{2\hbar^2}\int_{-\infty}^{\infty} \overline{[\mathcal{H}'(t'), [\mathcal{H}'(t'-\tau), \rho'(0)]]}\,d\tau$$

using the arguments of stationarity. We now use the replacement of $\bar{\rho}'(0)$ by $\bar{\rho}'(t)$ so we can write:

$$\dot{\bar{\rho}} = -\frac{1}{2\hbar^2}\int_{-\infty}^{\infty} \overline{[\mathcal{H}'(t'), [\mathcal{H}'(t'-\tau), \rho'(t)]]}\,d\tau$$

The error involved is of the order: $\frac{1}{\rho'(0)}\, t \Big/ \frac{d\rho'}{dt}$)

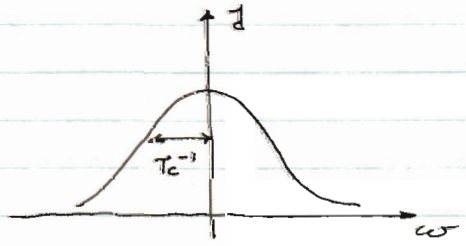
$$= \hbar^2 t \int_0^t \mathcal{H}'(t')\,\mathcal{H}'(t'-\tau)\,d\tau \simeq \hbar^2 |\mathcal{H}'|^2 t\,\tau_c \ll 1$$

Thus if $t \gg \tau_c$, $\mathcal{H}'$ must be very small as:

$$\hbar^{-1}\,|\mathcal{H}'|\,\tau_c \ll 1$$

which is a familiar form.

Consider a narrow band random process:



To make the above approximation, the frequency spectrum of the perturbing $\mathcal{H}$ must be more narrower than the above. We see that another factor that gives this requirement is:

$$\frac{|\rho_n|}{|\rho_{n-1}|} \simeq \hbar^{-1}\,|\mathcal{H}'|\,\tau_c \ll 1$$

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Recall:

$$\frac{d\rho'}{dt} = -\frac{1}{2\hbar^2}\int_{-\infty}^{\infty}\overline{[\mathcal{H}(t),[\mathcal{H}(t-\tau),\rho'(t)]]}\,d\tau$$

Taking matrix elements:

$$\frac{d\rho_{aa'}}{dt} = \frac{1}{2\hbar^2}\sum_{bb'}\rho_{bb'}\,e^{i\omega_{ab}t}\,e^{i\omega_{b'a'}t}$$

$$\cdot\left\{\int_{-\infty}^{\infty}\overline{\mathcal{H}_{ab}(t)\,\mathcal{H}_{b'a'}(t-\tau)}\,e^{-i\omega_{a'b'}\tau}\,d\tau\right.$$

$$\left.+\int_{-\infty}^{\infty}\overline{\mathcal{H}_{ab}(t-\tau)\,\mathcal{H}_{b'a'}(t)}\,e^{i\omega_{ab}\tau}\,d\tau\right.$$

Consider the Heisenberg time factors:

$$e^{i/\hbar\{E_a-E_b+E_{b'}-E_{a'}\}t}$$

We throw away non-secular perturbation terms for which $\hbar^{-1}(E_a-E_b+E_{b'}-E_{a'})\tau_c \gg 1$ and keep those secular perturbation for which the above is $\ll 1$.

Keep if: $a=a'$; $b=b'$

or: $a=b$; $a'=b'$

Only those relaxation are coupled together which have the same Heisenberg time factor. The equations now take the simplified form:

$$\dot{\rho}_{aa'} = \sum_{bb'} R_{aa'bb'}\,\rho_{bb'}$$

where R is a relaxation tensor. For the diagonal elements:

$$\dot{\rho}_{aa} = \sum_{b} R_{aabb}\,\rho_{bb}$$

We see that:

$$R_{aabb} = w_{ab}$$

which we have derived before.

This allows us to write:

$$\dot{\rho}_{aa} = \sum_{b \neq a} w_{ab}\, \rho_{bb} - \sum_{b'} w_{b'a}\, \rho_{aa}$$

If the perturbation is of the form $\mathcal{O} F(t)$ we have $w_{ba} = w_{ab}$.

Note if we have $\rho_{a\,b\neq a}(0) = 0$, the off-diagonal elements remain zero in time. Hence the diagonal elements describe the whole process under this initial condition.

This can be put in the form of a rate equation. Consider $\mathcal{N}$ atoms, $N_a = \mathcal{N} \rho_{aa}$. Then:

$$\frac{d N_a}{dt} = \sum_{b}' w_{ab} N_b - \left(\sum_{b}' w_{ab} \right) N_a$$

This is really $n-1$ independent equations. We look for solutions of the form: $N_a = N_a(0)\, e^{-\lambda t}$, etc. This leads to a secular determinant:

$$\begin{vmatrix} \lambda - \sum_{n}' w_{an} & -w_{ab} & -w_{ac} & \cdots \\ -w_{ab} & \lambda - \sum_{n}' w_{bn} & -w_{bc} & \cdots \\ \vdots & \vdots & \vdots & \end{vmatrix} = 0$$

whose solution gives the relaxation times.

Consider the off-diagonal elements. Take:

$$\dot{\rho}_{ab} = R_{ab\,ab}\, \rho_{ab}$$

Assume only off-diagonal elements. This leads to the result of last time:

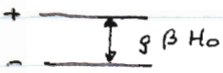
$$\dot{\rho}_{ab} = -\frac{1}{2} \left(\sum_{n}' w_{an} + \sum_{n}' w_{bn} \right) \rho_{ab}$$

$$= -\frac{1}{2} \left(\Gamma_{aa} + \Gamma_{bb} \right) \rho_{ab}$$

and: $$\left(\frac{d\rho}{dt}\right)^{\text{non-adiabatic}}_{\text{random perturbation}} = -\frac{1}{2}\left(\Gamma\rho + \rho\Gamma\right)$$

To get a feeling for the off-diagonal and diagonal relaxation processes, consider a Kramer's doublet.

$$\mathcal{H}_0 = -g\beta \underset{\sim}{S}\cdot\underset{\sim}{H}_0 = -g\beta H_0 S_z = \pm\frac{1}{2} g\beta H_0$$



Take for $\mathcal{H}(t) = 0\,F(t)$ where $F(t)$ is a stationary random process with a spectral density of:

$$g(\omega) = \int_{-\infty}^{\infty} \overline{F(t)\,F(t-\tau)}\, e^{i\omega\tau} d\tau$$

Then: $$W_{ab} = \hbar^{-2}\,|O_{ab}|^2\, g(\omega_{ab})$$

We may make the assignments: $\mathcal{H}(t) = g\beta(S_x H_x(t) + S_y H_y(t))$. Now:

$$\frac{dN^+}{dt} = W(N^- - N^+) = -\frac{dN^-}{dt}$$

or: $$\frac{d(N^+ - N^-)}{dt} = -2W(N^+ - N^-)$$

We see that the diagonal (or longitudinal) decay time is $T_1 = T_2 = \frac{1}{2W}$. Recall:

$$\langle M(t)\rangle = Tr\{M\rho(t)\}$$

Then: $$\frac{d}{dt}\langle M_z\rangle = \frac{1}{2} g\beta \underbrace{(\dot\rho_{++} - \dot\rho_{--})}_{\dot N^+ - \dot N^-}$$

$$= -\frac{1}{2} S\beta \frac{1}{T_1}(N^+ - N^-) = -\frac{\langle M_z\rangle}{T_1}$$

Thus is why T_1 is called the longitudinal decay time

For the off-diagonal:

$$\dot{\rho}^{na}_{+-} = -\frac{1}{2}(\omega_{+-} + \omega_{-+})\rho_{+-}$$

$$= -\omega\rho_{+-} = -\frac{\rho_{+-}}{2T_1}$$

For adiabatic:

$$\dot{\rho}_{+-} = -\frac{\rho_{+-}}{T_2}$$

where: $$\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2\text{'adiabatic}}$$

We find $T_2 = T_1$ if $\omega_0 \tau_c \ll 1$. This happens for random magnetic fields, but not for crystal field interaction.

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Recall:

$$\frac{dN_a}{dt} = \sum_{n'} w_{an'} N_{n'} - \left(\sum_{n'} w_{n'a} \right) N_a$$

$$N_a = p_{aa} n \quad ; \quad \sum N_a = n \quad ; \quad w_{an'} = w_{n'a}$$

The general solution is:

$$N_a = \sum_{i=1}^{m-1} c_i e^{-\lambda_i t} + c$$

We see that the steady state is $c = \frac{n}{m}$. However, this is the distribution at infinite temperature since:

$$N_a = n \frac{e^{-E_a/kT}}{\sum e^{-E_a/kT}}$$

This is a serious shortcoming of the above equation. This is avoided by treating the random perturbation as a quantum reservoir. This argument was that essentially used by Einstein in 1917. Because of thermodynamic conditions, the downward transition should exceed the upward transitions. After an infinite time, we need detailed balancing to get:

$$w_{an'} N_{n'} = w_{n'a} N_a$$

by induction. From statistical mechanics:

$$\frac{N_a}{N_{n'}} = e^{-(E_a - E_{n'})/kT} = \frac{w_{an'}}{w_{n'a}}$$

This makes:

$$c = n \frac{e^{-E_a/kT}}{\sum e^{-E_a/kT}}$$

This finishes diagonal relaxation.

We now return to off-diagonal relaxations:

$$\dot{\rho}'_{+-} = -\frac{\rho'_{+-}}{T_2}$$

where T_2 in the characteristic decay time of the off-diagonal elements.

Physical meaning of T_2. Consider a state of the form:

$$\psi = a_+ |+\rangle + a_- |-\rangle$$

which in not an eigenfunction of the energy Hamiltonian. This energy system may be taken to be a spin of ½ in a magnetic field. This system (2×2) uses the Pauli matrices regardless of if it is really a spin system. Here:

$$\mathcal{H}_0 = -g\beta \underset{\sim}{S}\cdot\underset{\sim}{H} = -g\beta S_z H_0 \quad ; \quad S_z = \tfrac{1}{2}\sigma_z$$
$$\underset{\sim}{M} = -g\beta \underset{\sim}{S}$$

——— $S_z = -\frac{1}{2}$

——— $S_z = +\frac{1}{2}$

Now our ψ is not an eigenfunction of the above H_0 but may be valid for M_x, M_y system. Consider:

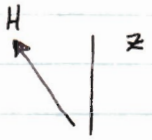
$$\langle M_x \rangle = (\psi^* | M_x | \psi) = g\beta \left[a_+^* \langle +1 + a_-^* \langle -| S_x | a_+ |+\rangle + a_- |-\rangle \right] = g\beta \left[\frac{1}{2} \left(\underbrace{a_+^* a_-}_{\rho_{+-}} + \underbrace{a_-^* a_+}_{\rho_{-+}} \right) \right]$$

Take $a_+ = a_- = \frac{1}{\sqrt{2}}$ which gives:

$$\langle M_x \rangle = \frac{g\beta}{2}$$

Thus our ψ describes the S_x state when we choose a_+, a_- properly. By taking a_+, a_- to be an imaginary ratio we get the S_y state.

Thus the off-diagonal elements allow us to describe states which are not eigenstates of the energy. Thus if we had:



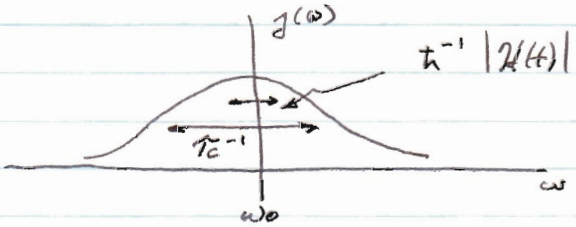
This system gives values for M_x, M_y. Now T_2 means that these values decay in amplitude with a characteristic time T_2. That is:

$$\frac{d}{dt}\langle M_x\rangle = \frac{d}{dt}\,Tr(\rho M_x) = Tr\, M_x \dot{\rho}$$

$$= -\frac{1}{T_2}\,Tr(M_x \rho) = -\frac{1}{T_2}\langle M_x\rangle$$

This is why T_2 is called the transverse decay time.

Let us now consider a purely periodic perturbation rather than a purely random one. We have considered:



Our treatment gave the relaxation behaviour when $\tau^{-1}|H(t)| \ll \tau_c^{-1}$

Clearly, a pure sinusoidal wave violates this criteria, because $g(\omega)$ becomes a δ function. Thus we start again from scratch. It will be convenient to use the field:

$$\left.\begin{aligned} H_x &= H_1 \cos\omega t \\ H_y &= H_1 \sin\omega t \end{aligned}\right\}\ \text{circularly polarized}$$

$$H_z = H_0$$

Now classically:

$$\frac{d\underset{\sim}{J}}{dt} = \underset{\sim}{M} \times \underset{\sim}{H} \quad ; \quad \underset{\sim}{J} = \frac{1}{\gamma}\underset{\sim}{M}$$

Hence: $\dot{\underset{\sim}{M}} = \gamma\,(\underset{\sim}{M} \times \underset{\sim}{H})$ and $\omega_0 = -\gamma H_0$

We now transform to a system rotating at ω, around the z axis (primed system). Then:

$$\frac{\partial \underset{\sim}{M}'}{\partial t} = \gamma(\underset{\sim}{M}' \times \underset{\sim}{H}') + \underset{\sim}{\omega} \times \underset{\sim}{M}' = \gamma(\underset{\sim}{M}' \times \underset{\sim}{H}_{eff})$$

(Note we have transformed away the time dependence)

where $H_{z\,eff} = H_0 + \frac{\omega}{\gamma}$

$H_{x\,eff} = H_{y\,eff} = H_1/\sqrt{2}$

The system will precess around $\underset{\sim}{H}_{eff}$ with a frequency:

$$\omega_{eff} = \gamma |\underset{\sim}{H}_{eff}| = \gamma\sqrt{H_1^2 + (H_0 + \frac{\omega}{\gamma})^2}$$

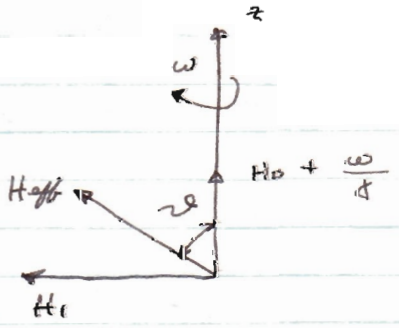
$$= \sqrt{\omega_1^2 + (\omega_0 + \omega)^2}$$

with: $M_z = M_0\{\cos^2\theta + \sin^2\theta \cos \omega_{eff} t\}$

$$= \{1 - 2\sin^2\theta \sin^2 \tfrac{1}{2}\omega_{eff} t\} M_0$$

$$M_x = M_0 \sin\theta\cos\theta(1 - \cos\omega_{eff} t)\sin\omega t + M_0 \sin^2\theta \sin\omega_{eff} t \cos\omega t$$

$M_y = M_x$ with $\sin\omega t \to \cos\omega t$, $\cos\omega t \to -\sin\omega t$



The quantum treatment gives the same answer.

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Recall:

$$\mathcal{H} = -g\beta\, \underline{S}\cdot\underline{H} = -\gamma\hbar\, \underline{S}\cdot\underline{H}$$

Last time we obtained the classical solution; we now consider the quantum result. Take:

$$H_z = H_0$$

$$H_x = H_1 \cos\omega t \;;\; H_y = H_1 \sin\omega t$$

Now:

$$i\hbar\dot{\psi} = -g\beta\left(S_z H_0 + \underbrace{H_1 S_x \cos\omega t}_{\frac{1}{2}H_1 S_+ e^{-i\omega t}} + \underbrace{H_1 S_y \sin\omega t}_{\frac{1}{2}H_1 S_- e^{i\omega t}}\right)\psi$$

Introduce: $\omega_0 = -\gamma H_0$; $\omega_1 = -\gamma H_1$; $\omega_{eff} = -\gamma H_{eff}$

Then:

$$i\dot{\psi} = \left(\omega_0 S_z + \tfrac{1}{2}\omega_1 S_+ e^{-i\omega t} + \tfrac{1}{2}\omega_1 S_- e^{i\omega t}\right)\psi$$

or:

$$i\dot{a}_+ = \tfrac{1}{2}\omega_0 a_+ + \tfrac{1}{2}\omega_1 e^{i\omega t} a_-$$

$$i\dot{a}_- = -\tfrac{1}{2}\omega_0 a_- + \tfrac{1}{2}\omega_1 e^{-i\omega t} a_+$$

The solution of these two equations is:

$$|a_-(t)|^2 = \rho_- = \sin^2\vartheta \sin^2 \tfrac{1}{2}\omega_{eff} t$$

with $a_+ = 1$ at $t = 0$.

$$\langle I_z\rangle = \tfrac{1}{2}(\rho_+ - \rho_-) = \tfrac{1}{2}(1 - 2\rho_-)$$

$$= \tfrac{1}{2}\left(\cos^2\vartheta + \sin^2\vartheta \cos\omega_{eff} t\right)$$

which is what we found classically. This is true also for:

$$\langle I_x\rangle = \tfrac{1}{2}\left(a_+^* a_- + a_-^* a_+\right)$$

and for $\langle I_y\rangle$.

To see that the classical and quantum solutions should be equal, consider:

$$i\hbar\,\dot{\underline{S}} = -[\mathcal{H}, \underline{S}] \quad ; \quad \mathcal{H} = -\gamma\hbar\,\underline{H}\cdot\underline{S}$$

Work out for one component:

$$\dot{S}_z = \gamma\underbrace{\{-H_x S_y + H_y S_x\}}_{(\underline{S}\times\underline{H})_z}$$

which is the classical equation of motion. To push the equality further, we do the quantum problem in the rotating coordinate system. Look for a transformation operator. Consider:

$$U = e^{-i\omega S_z t}$$

$$\psi = U\psi' \quad ; \quad \psi' = U^{-1}\psi$$

Then:

$$i\dot{\psi}' = U^{-1}\frac{\mathcal{H}}{\hbar}U\psi' \underbrace{-iU^{-1}\dot{U}\psi'}_{-\omega S_z\psi'}$$

Now:

$$U^{-1}S_z U = S_z$$
$$U^{-1}S_+ U = e^{i\omega t}S_+$$
$$U^{-1}S_- U = e^{-i\omega t}S_-$$

Then:

$$i\dot{\psi}' = \{(\omega_0-\omega)S_z + \omega_1 S_x\}\psi'$$

which corresponds to the Hamiltonian in the rotating coordinate system.
We could also make the transformation: $e^{-i\omega_1 S_y}$ which would give a Hamiltonian of the form:

$$i\dot{\psi}'' = \omega_{eff}S_z''\psi''$$

We could also apply: $e^{-i\omega_{eff}S_z' t}$, giving

$$i\dot{\psi}''' = (\omega_{eff} - \omega_{eff})\, S_z\, \psi''' = 0$$

so that ψ''' is time independent as suggested classically.

We should note that any two level quantum problem is related to the spin problem. Consider:

$$Q = \frac{1}{2} g_0 + \frac{1}{2} \underset{\sim}{g} \cdot \underset{\sim}{S}$$

as an operator of a two level system. The Hamiltonian will be:

$$\mathcal{H} = \frac{1}{2} E_0 - \gamma \hbar \underset{\sim}{H} \cdot \underset{\sim}{S}$$

where $\underset{\sim}{H}$ is a fictitious magnetic field and $\underset{\sim}{S}$ is a fictitious spin. Then the density matrix is:

$$\rho = \frac{1}{2} + \underset{\sim}{m} \cdot \underset{\sim}{S}$$

and $i\hbar \dot{\rho} = [\mathcal{H}, \rho]$ gives $\dot{\underset{\sim}{m}} = \gamma (\underset{\sim}{m} \times \underset{\sim}{H})$ which shows that any two level system behaves as a classical magnetic system.

Also:

$$\langle Q \rangle = \mathrm{Tr}\, \rho Q$$

so that:

$$\langle Q \rangle = \frac{1}{2} g_0 + \frac{1}{2} \underset{\sim}{m} \cdot \underset{\sim}{g}$$

We do not always have a rotating field, but sometimes have a linear field, in which we do not have a complete solution and we have to eliminate the non-secular terms that arise.

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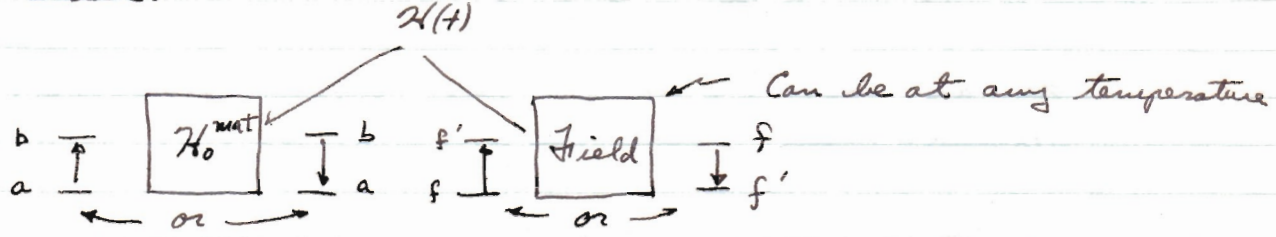
We now return to the random perturbation. We have seen that the diagonal elements relax to finite values which are given by the Boltzman factor.

We now will quantize the random perturbation. Consider:



The field could be a lattice phonon field.

Take the transitions:

$$P_{af';\,bf}(t) = \hbar^{-2} \int_0^t\int_0^t (bf|\mathcal{H}(t')|af')(af'|\mathcal{H}(t'')|bf) \cdot e^{-i\hbar^{-1}(E_b - E_a - E_{f'} + E_f)(t'-t'')}\, dt'\,dt''$$

Take $\mathcal{H}(t) = O\,F(t)$, then:

$$P_{af';\,bf}(t) = \hbar^{-2}\,|O_{ab}|^2 \int_0^t\int_0^t (bf|F(t')|af')(af'|F(t'')|bf)\cdots dt'\,dt''$$

Now F does not have an initial time dependence but does evolve with time according to the field Hamiltonian, that is:

$$F(t) = e^{i\hbar^{-1}\mathcal{H}t}\, F(0)\, e^{-i\hbar^{-1}\mathcal{H}t}$$

We now change to the τ coordinates system, taking the usual assumptions: We are interested now only in material system transitions. Therefore, we sum over all initial and final field states weighted according to a statistical density:

$$w_{ab} = \hbar^{-2}\,|O_{ab}|^2 \int_{-\infty}^{\infty} \sum_{ff'} \rho(f)\, \overline{(f|F(t)|f')(f'|F(t-\tau)|f)}\, e^{-i(\omega_{ab}-\omega_{ff'})\tau}\, d\tau$$

Take: $$p(f) = \frac{e^{-E_f/kT}}{\sum e^{-E_f/kT}}$$

Now the sum is really a trace, independent of a representation. This is really the quantum correlation function. Define:

$$P(\mathcal{H}) = \frac{e^{-\mathcal{H}/kT}}{Tr\left(e^{-\mathcal{H}/kT}\right)}$$

Then:

$$g(\tau) = Tr\left\{ F(t'+\tau)\, P(\mathcal{H})\, F(t') \right\}$$

so that we have the correlation function independent of the representation of the reservoir. We have used:

$$(f|F(t')|f')(f'|F(t+\tau)|f)$$

$$= e^{-i\hbar^{-1}(E_f - E_{f'})t'}\, e^{-i\hbar^{-1}(E_{f'} - E_f)(t'+\tau)}\, F_{ff'}(0)$$

Now:

$$J(\omega_{ab}) = \int_{-\infty}^{\infty} g(\tau)\, e^{i\omega_{ab}\tau}\, d\tau$$

And:

$$w_{ab} = \hbar^{-2}\, |O_{ab}|^2\, J(\omega_{ab})$$

But now we should have:

$$J(\omega_{ab}) = J(-\omega_{ab})\, e^{\hbar\omega_{ab}/kT}$$

Consider:

$$J(\omega) = \frac{1}{Tr\left(e^{-\mathcal{H}/kT}\right)} \int_{-\infty}^{\infty} \sum_{ff'} |(f|F|f')|^2\, e^{-E_f/kT}\, e^{i(\omega_{ff'} - \omega_{ab})\tau}\, d\tau$$

$$= \frac{1}{Tr\left(e^{-\mathcal{H}/kT}\right)} \sum_{f} |(f|F|f-\omega)|^2\, e^{-E_f/kT}$$

Now:

$$f(-\omega) = \frac{1}{\mathrm{Tr}(e^{-\mathcal{H}/kT})} \sum_{f''} |(f''|-\omega|F|f'')|^2 e^{-E_{f''}/kT} e^{\hbar\omega/kT}$$

QED.

This rule expresses the fact that the material system makes transition downward the preferred phenomena.

We now use this to explain spontaneous emission. Take $T=0$ or at least $h\nu \gg kT$. Then:

$$f(\omega_{ab}) = \frac{1}{\mathrm{Tr}(e^{-\mathcal{H}/kT})} \int_{-\infty}^{\infty} \sum_{d} |(0,0,\cdots \overset{n_\lambda}{|} F |0,0,\overset{n_\lambda}{1},\cdots)|^2 \cdot \underbrace{e^{i(\omega_\lambda - \omega_{ab})\tau} d\tau}_{\delta(\nu_\lambda - \nu_{ab})}$$

How many oscillators in a volume V? $\frac{\nu^2}{c^3} V d\Omega d\nu$ for one polarization.

The form of the interaction is:

$$\mathcal{H}_{1,0} = \frac{e}{mc} \left(\frac{2\pi\hbar c^2}{\omega_\lambda}\right)^{1/2} \underbrace{\int \psi_a^* (\vec{p}\cdot\hat{a}) e^{i\frac{\vec{k}}{\sim}\cdot\frac{\vec{r}}{\sim}} \psi_b \frac{dV}{V^{1/2}}}_{O_{ab}} \sqrt{1} \frac{1}{V^{1/2}}$$

Then:

$$W_{ab}^{\text{spont.}} d\Omega = \hbar^{-2} |O_{ab}|^2 \int_0^\infty \frac{\nu^2 d\Omega}{c^3} \frac{2\pi\hbar c^2}{(2\pi\nu)^2} \left(\frac{e}{mc}\right)^2 \delta(\nu - \nu_{ab}) d\nu$$

$$= \hbar^{-2} |O_{ab}|^2 \frac{\nu}{\hbar c} \left(\frac{e}{mc}\right)^2 d\Omega$$

for one polarization.

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Recall:

$$P_{af';bf} = \hbar^{-2} \int_0^t \int_0^t (bf|H_{int}|af')(af'|H_{int}|bf)\, e^{-i\hbar^{-1}(E_a - E_b - E_f + E_{f'})(t'-t'')}\, dt'\, dt''$$

where:

$$H_{int} = O_{op}^{mat}\, F_{op}^{reservoir}(t)$$

$$F(t) = e^{i\hbar^{-1}\mathcal{H}t}\, F(0)\, e^{-i\hbar^{-1}\mathcal{H}t}$$

obtaining:

$$w_{ab} = \hbar^{-2} |O_{ab}|^2 \int_{-\infty}^{\infty} (f|F(t')|f')(f'|F(t'')|f)\, e^{-i\omega_{ab}\tau}\, d\tau$$

We now take the reservoir to be the vacuum states of the electromagnetic field at $T=0$. This will give us spontaneous emission. Consider:

$$\frac{P}{t} = w_{af';bf} = \hbar^{-2} |O_{ab}|^2 \int_{-\infty}^{\infty} F_{ff'}(0)\, F_{f'f}(0)\, e^{-i(\omega_{ab} - \omega_{ff'})\tau}\, d\tau$$

Then:

$$w_{af;bf}^{sp.} = \hbar^{-2} |O_{ab}|^2 \int_{-\infty}^{\infty} \sum_{f'} |F_{ff'}|^2\, e^{-i(\omega_{ab} - \omega_{ff'})\tau}\, d\tau$$

$\sum_{f'}$ goes into a summation of oscillators and can be replaced by an integral.

Take:

$$O_{ab} = \int \psi_a^* (\vec{p}\cdot\hat{a})\, e^{i\vec{k}\cdot\vec{r}}\, \psi_b\, dV$$

$$F = \frac{e}{mc} \left(\frac{2\pi c^2 \hbar}{\omega_\lambda}\right)^{1/2} \frac{1}{V^{1/2}} \sqrt{n_\lambda + 1}$$

$$w_{ab}^{sp.\ per\ unit\ d\Omega}\, d\Omega = \hbar^{-2} |O_{ab}|^2 \int_0^\infty \left(\frac{e}{mc}\right)^2 \frac{2\pi c^2 \hbar}{\omega_\lambda}\, \delta(\nu_{ab} - \nu_\lambda)\, \frac{\nu_\lambda^2\, d\Omega\, d\nu_\lambda}{c^3}$$

$$= |O_{ab}|^2 \frac{2\nu_{ab}}{\hbar c} \left(\frac{e}{mc}\right)^2 d\Omega$$

We assume that $e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}$ varies slowly over the volume concerned. Use:

$$\left(\frac{\underset{\sim}{p}}{m}\right)_{ab} = (\underset{\sim}{v})_{ab} = 2\pi i\, \nu_{ab}\, \underset{\sim}{r}$$

Then:

$$|O_{ab}|^2 = 4\pi^2 \nu_{ab}^2 m^2 |x_{ab}|^2 \begin{cases} \cos^2 \textcircled{H} \\ \cos^2 \textcircled{H}' \end{cases}$$

Call $\textcircled{H}$ the angle between $\bar{a}$ and x direction

" $\textcircled{H}'$ " " " " other polarization " " "

We have $\cos^2 \textcircled{H} + \cos^2 \textcircled{H}' = \sin^2 \vartheta$

Then the transition probability regardless of polarization in a solid angle $d\Omega$ is:

$$w_{ab}^{ap} = \int_{-\pi}^{\pi} \int_{0}^{2\pi} \frac{4\pi^2 \nu_{ab}^3}{\hbar^2 c^3} e^2 |x_{ab}|^2 \sin^2\vartheta \sin\vartheta\, d\vartheta\, d\varphi$$

Then:

$2\pi\hbar\nu_{ab} W_{ab}^{spont.}$ = total spontaneous emission power

$$= \frac{64\pi^4 \nu_{ab}^4}{3c^3} e^2 |x_{ab}|^2$$

Consider finite temperatures. All we have to do is replace 1 by $\langle n_a \rangle + 1$ due to the occurence of $p(n_a)(n_a+1)$. Then, because $\bar{n}_a$ corresponds to an initial pumping of the system:

Total spontaneous and stimulated emissive power

$$= \frac{64\pi^4 \nu_{ab}^4}{3c^3} e^2 |x_{ab}|^2 (\bar{n}_a + 1) \qquad (w_{a\to b})$$

The total absorbed power is this with $\bar{n}_b + 1 \to \bar{n}_a$ with $w_{b\to a}$.

The $\bar{n}_\lambda$ is given by Planck's Formula:

$$\bar{n}_\lambda = \frac{1}{e^{\frac{\hbar\omega_\lambda}{kT}} + (-1)} = \frac{1}{e^{\frac{\hbar\omega_\lambda}{kT}} - 1}$$

Now in the steady state:

$$N_a \, w_{b\leftarrow a} = N_b \, w_{a\leftarrow b}$$

so that:

$$\frac{N_b}{N_a} = \frac{w_{b\leftarrow a}}{w_{a\leftarrow b}} = \frac{\bar{n}_\lambda}{\bar{n}_\lambda + 1} = e^{-\frac{\hbar\omega_{ab}}{kT}}$$

We now consider the line broadening. Recall:

$$\frac{1}{T_2} = \frac{1}{2}\left(\Gamma_{aa} + \Gamma_{bb}\right)$$

Now $\Gamma_{bb} \propto \bar{n} + 1$; $\Gamma_{aa} \propto \bar{n}$ at finite temperatures so that:

$$\left(\frac{1}{T_2}\right)_T = (2\bar{n} + 1)\left(\frac{1}{T_2}\right)_0$$

We consider a damped material system (damped by all different field oscillators) in interaction with one particular mode of the radiation field. In this case:

$$P_{a\, n_\lambda+1,\, b\, n_\lambda} = \hbar^{-2} \left| H_{a\, n_\lambda+1,\, b\, n_\lambda} \right|^2 \int_0^t \int_0^t \langle a(t'') | \; |b(t'')\rangle\langle b(t'')| \; |a(t')\rangle \cdot e^{i(\omega_{ab} - \omega_\lambda)(t' - t'')} \, dt' \, dt''$$

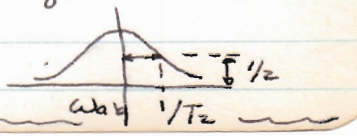
Using the fact that $a(t) = a(0)\, e^{-\frac{\Gamma_{aa}}{2}t}$ even though we have removed one mode from the ensemble:
Another way: $\overline{a(t)\, a^*(t'')} = |a(0)|^2 e^{-\frac{\Gamma_{aa}}{2}|\tau|}$:

Thus:

$$w_{a\, n_\lambda+1,\, b\, n_\lambda} = \hbar^2 \left| H_{a\, n_\lambda+1,\, b\, n_\lambda} \right|^2 \int_{-\infty}^{\infty} e^{-\frac{\Gamma_{aa}}{2}|\tau|} \; e^{-\frac{\Gamma_{bb}}{2}|\tau|} \; e^{-i(\omega_{ab} - \omega_a)\tau} \, d\tau$$

$$= \hbar^{-2} \left| H_{a\, n_\lambda+1,\, b\, n_\lambda} \right|^2 \left\{ \frac{2T_2}{(\omega_{ab} - \omega_\lambda)^2 T_2^{-2} + 1} \right\}$$

Lorentzian:



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Recall:

$$W_{n_a+1,\,n_b} = \hbar^{-2} \left| \mathcal{H}_{ab}\, n_a+1,\, b\, n_b \right|^2 \rho \int_{-\infty}^{\infty} a(t')\, a^*(t'+\tau)\, b^*(t')\, b(t'+\tau)$$

$$e^{i(\omega_{ba}-\omega_a)\tau}\, d\tau$$

An application would be for a paramagnetic salt in a cavity that sustains one mode at the resonance of ω_{ab}. How do we apply this equation when it was derived for random time perturbations? The answer is that the random time perturbations come from the lattice and this is what the a and b above refer to. However, this would not apply to a single atom in the cavity. Here what has essentially been done is to consider a purely periodic perturbation and a random perturbation at the same time by picking out a single mode of the random perturbation. From previous work ($a\,a^* \to e^{-\frac{\Gamma_{aa}}{2}|\tau|}$), the above integral becomes:

$$\int_0^{\infty} e^{-\frac{\Gamma_{aa}}{2}\tau}\, e^{-\frac{\Gamma_{bb}}{2}\tau}\, e^{i(\omega_{ba}-\omega_a)\tau}\, d\tau$$

$$+ \int_0^{\infty} e^{-\frac{\Gamma_{aa}}{2}\tau}\, e^{-\frac{\Gamma_{bb}}{2}\tau}\, e^{-i(\omega_{ba}-\omega_a)\tau}\, d\tau$$

$$= \frac{1}{i(\omega_{ba}-\omega_a)+T_2^{-1}} + \frac{1}{-i(\omega_{ba}-\omega_a)+T_2^{-1}} \quad ; \quad T_2^{-1} = \frac{\Gamma_{aa}+\Gamma_{bb}}{2}$$

This leads to a spectral density of:

$$g(\nu_a) = \frac{2T_2}{(\omega_{ba}-\omega_a)^2 T_2^2 + 1}$$

(This is the line shape function and is the Fourier transform of the off-diagonal density matrix element)

This line shape function is the same for both stimulated emission and absorption.

We now rederive the same result concentrating on the material system and hope to arrive at a point where we have the macroscopic Maxwellian material response functions. The model is the same as above. Consider:

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho] + \mathcal{W}_{\text{damping}}$$

Take as initial conditions:

$$\left.\begin{aligned} &\rho_{aa}\ ;\ \rho_{bb}\ ;\ \cdots \\ &\rho_{ab}(t=0) = 0 \end{aligned}\right\} \text{ in absence of periodic perturbations}$$

We are taking the ρ's to be taken over a statistical ensemble so that for initial conditions we have:

$$\rho_{aa} = \frac{e^{W_a/kT}}{\sum e^{W_a/kT}}\ \rho_{bb}$$

We take the perturbation to be a precessing magnetic field:

$$\mathcal{H}_{ab} e^{i\omega t} + \mathcal{H}_{ab}^{*} e^{-i\omega t}$$

giving:

$$i\hbar\,\dot{\rho}_{ab} = (\mathcal{H}_{ab}\rho_{bb} - \mathcal{H}_{ab}\rho_{aa}) + \underbrace{(\mathcal{H}_{aa} - \mathcal{H}_{bb})}_{\hbar\omega_{ab}}\rho_{ab}$$

$$- i\hbar\, T_2^{ab\,-1}\,\rho_{ab}$$

We consider only two levels and take a periodic solution, finding:

$$\rho_{ab} = \frac{\hbar^{-1}\mathcal{H}_{ab}}{-\omega + \omega_{ba} + i T_2^{-1}}\ (\rho_{bb} - \rho_{aa}) \quad \left\{ \text{similar for } \rho_{ba} \right\}$$

Take for the precessing field:

$$\mathcal{H}_{ab} = -\frac{1}{2}\gamma\hbar\, S^{+}_{ab}\,(H_x - i H_y)$$

Consider:

$$\langle M_x - i M_y \rangle = 2\cdot\frac{1}{2}\gamma\hbar\langle S_- \rangle = 2\cdot\frac{1}{2}\gamma\hbar\, Tr\,(S_- \rho)$$

$$= \frac{-\frac{2\cdot 1}{4}\gamma^2\hbar\,(H_x - i H_y)\,e^{i\omega t}}{-\omega + \omega_{ba} + i T_2^{-1}}\,(\rho_{bb} - \rho_{aa})$$

Now we can define:

$$\chi' - i\chi'' = \frac{\langle M_x - i M_y \rangle}{H_x - i H_y}$$

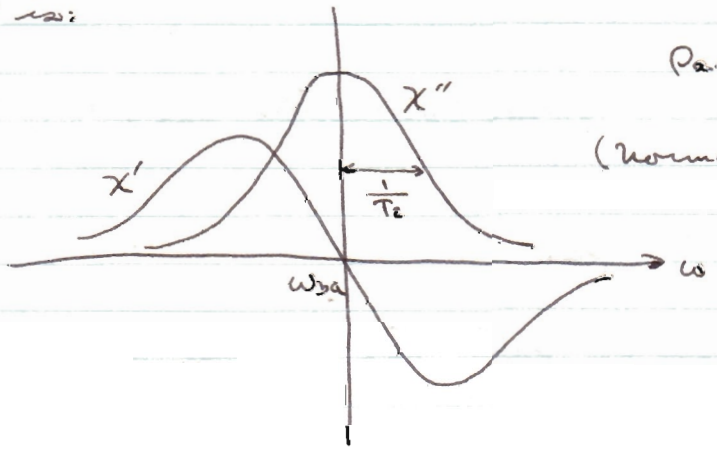
and thus obtain the susceptibility from the quantum mechanics.

$$\chi'' = \frac{\frac{1}{2}\gamma^2\hbar\, T_2}{1 + (\omega_{ba} - \omega)^2 T_2^2}\,(\rho_{aa} - \rho_{bb})$$

$$\chi' = (\omega_{ba} - \omega)\, T_2\, \chi''$$

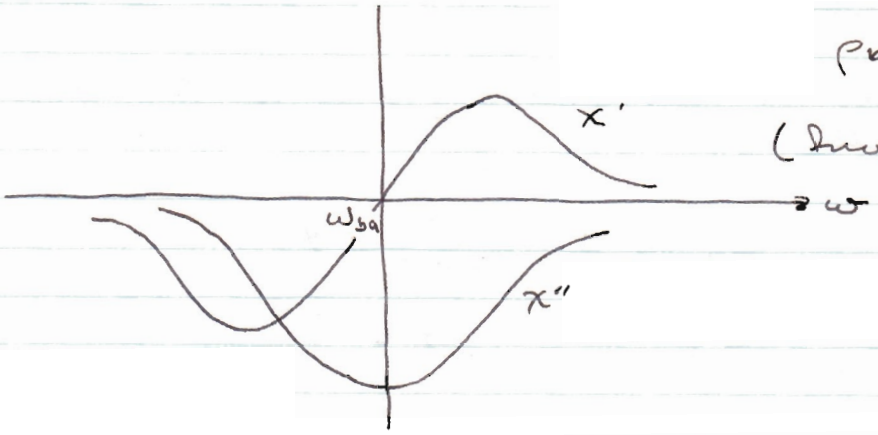
Note: $\frac{1}{2}\gamma\hbar\,(\rho_{aa} - \rho_{bb}) = \langle M_z \rangle$

The plot is:



$\rho_{aa} > \rho_{bb}$ { — b, — a }

(normal)



$\rho_{bb} > \rho_{aa}$ $\left\{ \begin{matrix} -b \\ -a \end{matrix} \right\}$

(Inverted)

This is the condition of maser action as the incoming wave is amplified by the negative χ''. Although we have not considered spontaneous emission, it will appear as noise.

The above equations are known as the Bloch equations, which be derived from:

$$\dot{M}_{x,y} = \gamma (\underset{\sim}{M} \times \underset{\sim}{H})_{x,y} - \frac{M_{x,y}}{T_2}$$

a classical equation which be assumed to be damped.

We can get the result for the linear polarization by recalling that a linearly polarized wave may be resolved into two counter-rotating circularly polarized waves. Actually, this leads to susceptibilities that are 1/2 of those for circularly polarized waves, except for $\omega = 0$. The above of the equations are the form of the Kramers-Heisenberg dispersion formula for magnetic dipole radiation.

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Consider the following form of the interaction Hamiltonian:

$$\mathcal{H} = -\frac{e}{mc}\,\underset{\sim}{p}\cdot\underset{\sim}{A} + \frac{e^2}{2mc^2}\,\underset{\sim}{A}\cdot\underset{\sim}{A}$$

$$\underset{\sim}{A} = \mathrm{Re}\left[\hat{a}\,(2\pi c^2)^{-1/2}\,q\,e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}-i\omega t}\right]$$

We will calculate the expectation value of the current $\underset{\sim}{j} = e\underset{\sim}{v}$ in order to eventually obtain the conductivity. Now:

$$\underset{\sim}{v} = \frac{1}{m}\left(\underset{\sim}{p} - \frac{e}{c}\underset{\sim}{A}\right) \qquad \text{(no DC vector potential)}$$

Recall:

$$\rho_{nn'} = \frac{\hbar^{-1}\,\frac{e}{mc}\,q\,(2\pi c^2)^{1/2}\,\langle n|\underset{\sim}{p}\cdot\hat{a}\,e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}|n'\rangle}{+\omega + \omega_{n'n} + i\,T_2^{-1}}$$

$$\cdot\,(\rho_{n'n'} - \rho_{nn})\,e^{-i\omega t} + \frac{\cdots\cdots}{-\omega + \omega_{n'n} + i\,T_2^{-1}}\,e^{i\omega t}$$

Take the expectation of $\underset{\sim}{j}$ in some direction, say $\hat{a}'$ not necessarily in the same direction as $\hat{a}$, by the usual trace procedure. Consider $\underset{\sim}{v} = \frac{\underset{\sim}{p}}{m}$ for the moment.

$$\langle\underset{\sim}{j}\cdot\hat{a}'\rangle = \mathrm{Re}\,\frac{e^2}{m^2c}\,q\,(2\pi c^2)^{-1/2}\sum_{nn'}\left\{e^{-i\omega t}\left[\frac{\langle n'|\underset{\sim}{p}\cdot\hat{a}'|n\rangle\langle n|\underset{\sim}{p}\cdot\hat{a}\,e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}|n'\rangle}{\hbar\,(\omega + \omega_{n'n} + i\,T_2^{-1})}\right.\right.$$

$$\left.\left. + \frac{\langle n(\underset{\sim}{p}\cdot\hat{a}')n'\rangle\langle n'|\underset{\sim}{p}\cdot\hat{a}\,e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}|n\rangle}{\hbar\,(-\omega + \omega_{n'n} + i\,T_2^{-1})}\right]\rho_{nn}\right\}$$

We will only consider terms linear in $\underset{\sim}{A}$ and at the same frequency. Consider:

$$\left\langle -\frac{e^2}{mc}\,\underset{\sim}{A}\right\rangle$$

This adds the term:

$$-\mathrm{Re}\; q\,(2\pi c^2)^{-1/2}\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}-i\omega t} \sum_n \rho_{nn} \left\langle \frac{e^2}{mc}\underset{\sim}{A} \right\rangle$$

This last term is important as it is the only term left in the case of a electron gas. We have now really found the conductivity tensor because:

$$\underset{\sim}{E} = -\frac{1}{c}\frac{\partial \underset{\sim}{A}}{\partial t} = \mathrm{Re}\left[\frac{i\omega}{c}\hat{a}\,(2\pi c^2)^{-1/2} q\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}-i\omega t}\right]$$

$$= \underset{\sim}{E_0}\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}-i\omega t}$$

Then we deduce:

$$\sigma_{\hat{a},\hat{a}'} = \frac{e^2}{m^2 c\,\frac{i\omega}{c}} \sum_{nn'} \rho_{nn'} \{\cdots\}$$

This conductivity tensor also contains all the magnetic information about the medium. The above current density is from:

$$\nabla \times \underset{\sim}{H} = \frac{4\pi}{c}\underset{\sim}{j}_{mat} + \frac{1}{c}\frac{\partial \underset{\sim}{E}}{\partial t}$$

For the electric dipole case, $e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \approx 1$ with $\underset{\sim}{j} = \frac{\partial \underset{\sim}{P}}{\partial t} = -i\omega \underset{\sim}{P}$, giving:

$$\frac{\epsilon_{\hat{a}\cdot\hat{a}}-1}{4\pi} = \frac{\langle \underset{\sim}{P}\cdot\hat{a}'\rangle}{E_0} = \frac{e^2}{m^2 c\,\frac{\omega^2}{c}} \sum_{nn'} \rho_{nn} \{\cdots\} = \chi_{\hat{a}\hat{a}'}$$

For an isotropic medium:

$$\chi = \frac{e^2}{m^2\omega^2} \sum_{nn'} \rho_{nn} \left\{ \frac{(p_x)_{n'n}(p_x)_{nn'}}{\hbar\left(\omega + \omega_{n'n} + i\Gamma_{n'n}^{-1}\right)} + \frac{(p_x)_{nn'}(p_x)_{n'n}}{\hbar\left(-\omega + \omega_{n'n} + i\Gamma_{n'n}^{-1}\right)} \right\}$$

$$- \frac{e^2}{mn\omega^2} \sum_n \rho_{nn}$$

Now consider the case of no damping: This gives:

$$\chi = \frac{e^2}{\omega^2} \sum_{nn'} \rho_{nn} \left[\frac{|x_{nn'}|^2 \omega_{nn'}^2}{\hbar(\omega + \omega_{n'n})} + \frac{|x_{nn'}|^2 \omega_{nn'}^2}{\hbar(-\omega + \omega_{n'n})} \right]$$

$$- \frac{e^2}{m\omega^2} \sum_n \rho_{nn}$$

Using $[px, x] = \frac{\hbar}{i}$ and taking matrix elements in any given representation gives:

$$\sum_{n'} 2\omega_{n'n} |x_{nn'}|^2 = \hbar m^{-1}$$

Substitution into χ gives:

$$\chi = \frac{e^2}{\omega^2} \sum_{nn'} \rho_{nn} \left\{ \frac{|x_{nn'}|^2 \omega_{n'n}^2 - \omega_{n'n} |x_{nn'}|^2 (\omega + \omega_{n'n})}{\hbar(\omega + \omega_{n'n})} \right.$$

$$\left. + \frac{|x_{n'n}|^2 \omega_{n'n}^2 - \omega_{n'n}(-\omega + \omega_{n'n})}{\hbar(-\omega + \omega_{n'n})} \right\}$$

or:

$$\chi = \frac{e^2}{\omega} \sum_{nn'} \rho_{nn} \left\{ \frac{-\omega_{n'n} |x_{n'n}|^2}{\hbar(\omega + \omega_{n'n})} + \frac{|x_{n'n}|^2 \omega_{n'n}}{\hbar(-\omega + \omega_{n'n})} \right\}$$

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Recall that we had derived the Kramers - Kronig dispersion relation:

$$\chi = \sum_{nn'}' \frac{|e X_{nn'}|^2}{\hbar^2 (\omega^2 - \omega_{nn'}^2)} \rho_{nn} \eta_0$$

where η_0 is the number of particles per unit volume. We can get the complex susceptibility by replacing in the original formula, $\omega_{nn'} \to \omega_{nn'} + i\Gamma_2^{-1}$, obtaining:

$$\chi' + i\chi'' = \frac{e^2}{\omega} \sum_{nn'} \left\{ \frac{|X_{n'n}|^2 \omega}{\hbar(-\omega + \omega_{nn'} + i\Gamma_{2\,nn'}^{-1})} + \frac{|X_{n'n}|^2 \omega}{\hbar(\omega + \omega_{nn'} + i\Gamma_{2\,n'n}^{-1})} \right\} \rho_{nn} \eta_0$$

This is similar to the spin resonance case. The above, keeping only resonant terms, for the case of two levels reduces to:

$$\chi' + i\chi'' = \frac{|P_x^{ab}|^2}{\hbar(-\omega + \omega_{ba} + i\Gamma_2^{-1})} (\rho_{aa} - \rho_{bb}) \eta_0$$

which we compare to:

$$\chi' - i\chi'' = \frac{\frac{1}{4}\gamma^2\hbar^2 S_{ab}^+ S_{ba}}{\hbar(-\omega + \omega_{ba} + i\Gamma_2^{-1})} (\rho_{aa} - \rho_{bb})$$

$$= \frac{|M_x^{ab}|^2}{\hbar(-\omega + \omega_{ba} + i\Gamma_2^{-1})} (\rho_{aa} - \rho_{bb})$$

We see that the above results could be given by:

$$\mathcal{H} = -\underset{\sim}{M} \cdot \underset{\sim}{H_0}$$

and

$$\mathcal{H}' = -e\underset{\sim}{r} \cdot \underset{\sim}{E}$$

$\mathcal{H}'$ is equivalent to: $\mathcal{H} = -\frac{e}{c}\underset{\sim}{p}\cdot\underset{\sim}{A} + \frac{e^2}{2mc^2}\underset{\sim}{A}\cdot\underset{\sim}{A}$

(providing that the spatial dependence of $\underset{\sim}{A}$ is negligible) in that it leads to above results. We now show this: (M. Goeppert-Mayer, Hand. d. Phys. 24, 273 (1931)): Recall.

$$\mathcal{H} = \frac{\underset{\sim}{p}^2}{2m} + V(\underset{\sim}{r}) - \frac{e}{mc}\underset{\sim}{p}\cdot\underset{\sim}{A}(\underset{\sim}{r},t) + \frac{e^2}{2mc^2}\underset{\sim}{A}^2(\underset{\sim}{r},t)$$

$$\mathcal{L} = \frac{1}{2}m\dot{\underset{\sim}{r}}^2 - V(\underset{\sim}{r}) + \frac{e}{c}\dot{\underset{\sim}{r}}\cdot\underset{\sim}{A}(\underset{\sim}{r},t)$$

Now we can add any time derivative to the Lagrangian, say $\frac{d}{dt}(-\underset{\sim}{r}\cdot\underset{\sim}{A})$, forming an equivalent Lagrangian:

$$\mathcal{L}' = \frac{1}{2}m\dot{\underset{\sim}{r}}^2 - V(\underset{\sim}{r}) - \frac{e}{c}\underset{\sim}{r}\cdot\frac{d}{dt}\underset{\sim}{A}(\underset{\sim}{r}t)$$

$$\mathcal{H}' = \frac{\underset{\sim}{p}^2}{2m} + V(\underset{\sim}{r}) + \frac{e}{c}\underset{\sim}{r}\cdot\frac{d\underset{\sim}{A}(\underset{\sim}{r},t)}{dt}$$

Now, if we can put $\frac{d}{dt} \to \frac{\partial}{\partial t}$, we can use:

$$\underset{\sim}{E} = -\frac{1}{c}\frac{\partial\underset{\sim}{A}}{\partial t}$$

and we can do just this if the spatial dependence is negligible ($\frac{d}{dt} = \frac{\partial}{\partial t} + \nabla\cdot$).

Now, can we make a transformation from the vector potential Hamiltonian to one with a multipole expansion form? This has been done by: J. Fenton, Can. J. Phys. 41, 12 (1963). That is, we can write:

$$\mathcal{H}' = -e\underset{\sim}{r}\cdot\underset{\sim}{E} - \underset{\sim}{M}\cdot\underset{\sim}{H} - \underset{\sim}{Q}\cdot\nabla\underset{\sim}{E} + \cdots$$

All of our above calculation of response functions have assumed a non-dense medium so that there is sense in writing $\mathcal{H}_0$. Now, in a dense media, we must use the Lorentz-Lorenz

model of an effective field due to neighboring atomic systems. Consider:

$$\underset{\sim}{E}_{loc} = \underset{\sim}{E}_{ins} + \underset{\sim}{\underset{\sim}{L}} \cdot \underset{\sim}{P} \quad ; \quad \underset{\sim}{P} = \underset{\sim}{\underset{\sim}{\chi}} \cdot \underset{\sim}{E}_{loc}$$

This is similar to the Weiss model. For an isotropic structure, Lorentz showed:

$$\underset{\sim}{\underset{\sim}{L}} = \frac{4\pi}{3}$$

so that:

$$\underset{\sim}{P} = \frac{\chi}{1 - \frac{4\pi}{3}\chi} \underset{\sim}{E} \quad ; \ \chi \text{ is the atomic susceptibility.}$$

Now: $\underset{\sim}{D} = \underset{\sim}{\underset{\sim}{\epsilon}} \underset{\sim}{E} = \underset{\sim}{E} + 4\pi \underset{\sim}{P}$, hence:

$$\frac{\epsilon - 1}{4\pi} = \frac{\chi}{1 - \frac{4\pi}{3}\chi} \qquad \text{(Lorentz - Lorenz Relation)}$$

Now, $\underset{\sim}{E}_{inside}$ for an ellipsoidal body is given by:

$$\underset{\sim}{E}_{inside} = \underset{\sim}{E}_{outside} - \underbrace{\underset{\sim}{\underset{\sim}{N}} \cdot \underset{\sim}{P}}_{\text{depolarization}}$$

If the body is a small sphere, then the depolarization field cancels the internal field and we have that the local field equals the outside field.

Consider now Maxwell's equations:

$$\nabla \times \underset{\sim}{E}_{vac} = -\frac{1}{c}\frac{\partial \underset{\sim}{H}_{vac}}{\partial t}$$

$$\nabla \times \underset{\sim}{H}_{vac} = \frac{1}{c}\frac{\partial \underset{\sim}{E}_{vac}}{\partial t} + \frac{4\pi}{c}\underset{\sim}{j}_{mat}$$

$$\nabla \times \nabla \times \underset{\sim}{E}_{vac} + \frac{1}{c^2}\frac{\partial^2 \underset{\sim}{E}_{vac}}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial \underset{\sim}{j}}{\partial t}$$

$\underset{\sim}{j}$ is approximately:

$$\underset{\sim}{j} = \frac{\partial \underset{\sim}{P}}{\partial t} + \frac{1}{c}\nabla \times \underset{\sim}{M} - \nabla \cdot \frac{\partial \underset{\sim}{\underset{\sim}{Q}}}{\partial t} + \cdots$$

We can envision an effective conductivity, or, an effective dipole moment (properly retarded):

$$\underset{\sim}{j} = \frac{\partial \underset{\sim}{P}}{\partial t}$$

Now use $\underset{\sim}{P} = \frac{\epsilon - 1}{4\pi}\underset{\sim}{E}$ and we get the usual wave equation:

$$\nabla \times \nabla \times \underset{\sim}{E} + \frac{\epsilon}{c^2}\frac{\partial^2 \underset{\sim}{E}}{\partial t^2} = 0$$

whose solution is:

$$\underset{\sim}{E} = \hat{a} E_0 e^{i \underset{\sim}{k} \cdot \underset{\sim}{x} - i\omega t} \quad ; \quad \hat{a} \cdot \underset{\sim}{k} = 0$$

where: $|\underset{\sim}{k}| = \frac{\omega}{c}\epsilon^{1/2} = \frac{\omega}{c} n (1 + i\kappa)$

where κ is the attenuation ~~index~~. We see that the wave attenuates as:

$$E \propto e^{-\frac{\omega n \kappa}{c}|z|}$$

The attenuation coefficient comes from $EE^* \propto e^{-\frac{2\omega n \kappa}{c}|z|} = e^{-\alpha |z|}$ so that:

$$\alpha = \frac{2\omega n \kappa}{c} = \frac{4\pi}{\lambda} n$$

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Erratta: $\underset{\sim}{j}_{mat} = \frac{\partial \underset{\sim}{P}}{\partial t} + c \nabla \times \underset{\sim}{M} - \nabla \frac{\partial Q}{\partial t}$

Recall:

$$\nabla \times E_{vac} = -\frac{1}{c} \frac{\partial B_{vac}}{\partial t}$$

$$\nabla \times B_{vac} = \frac{1}{c} \frac{\partial E_{vac}}{\partial t} + 4\pi j_{mat}$$

$$\nabla \times H_{mat} = \frac{1}{c} \frac{\partial D_{mat}}{\partial t}$$

$H_{mat} = B_{vac} - 4\pi M_{mat}$

$D_{mat} = E_{vac} + 4\pi P_{mat}$

Recall the susceptibility of a two level magnetic system:

$$\chi' - i\chi'' = \frac{|P^{ab}|^2 n}{\hbar(-\omega + \omega_{ba} + i T_2^{-1})} (\rho_{aa} - \rho_{bb})$$

Why does an ensemble of atoms propagate a plane wave and can be described by a response function, whereas, we get dipole scattering off a single atom?

The reason is that we have assumed in taking the ensemble average that each element of volume has the same probability of containing an atom. If one went thru the mathematics of adding all the wavelets from each atom one would find destructive interference everywhere except in the direction of the incident wave.

This allows us to calculate response function with QM and then use Maxwell's equations for further discussion of coherent radiation.

How does the above describe a solid? If the wavelength of I_0 is larger than the interatomic distance, we are all right, because we have the same situation as a gas.

In conclusion, the ensemble will only add or subtract from the incident wave in the case of a perfect crystal or a uniform density gas.

Kramers - Kronig Relations (Causality) (1926)

This is useful when we have a system that obeys the "Golden Rule" of absorption:

$$\hbar^{-2} \underbrace{|H_{ab}|^2}_{|P_{ab}|^2 |E|^2} g(\nu) (N_a - N_b) \hbar\nu = \tfrac{1}{2} 2\pi\nu \chi'' |E|^2$$

Take a Lorentzian line shape: $g(\nu) = \dfrac{2T_2}{(-\omega+\omega_{ba})^2 T_2^2 + 1}$

We can now use the KK relation to get χ'

The KK relations are a general property of linear, stationary, finite systems. Consider the unit response $f(t)$ for a unit pulse $\delta(t)$. Also, we impose the causality condition $f(t) = 0$, $t < 0$.

Consider an ensemble of δ functions $S(t)$ for a stationary input signal. Then the stationary output, which also assumes linearity, is:

$$R(t) = \int_{-\infty}^{\infty} S(t') f(t-t')\, dt'$$

Take: $S(t) = E_0 e^{i\omega t}$:

$$R(t) = E_0 e^{i\omega t} \underbrace{\int_{-\infty}^{\infty} e^{i\omega(t'-t)} f(t'-t)\, dt'}_{\int_{-\infty}^{\infty} e^{i\omega t''} f(t'')\, dt'' \;=\; \text{independent of time}}$$

Now, if we take an electromagnetic system, we immediately see:

$$\chi(\omega) = \int_0^\infty e^{-i\omega t''} f(t'')\, dt''$$

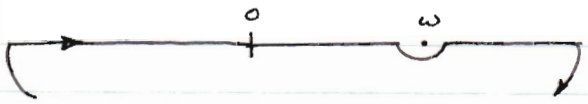
$$\chi'(\omega) = \int_0^\infty \cos \omega t \;\; f(t)\, dt$$

$$\chi''(\omega) = \int_0^\infty \sin \omega t \;\; f(t)\, dt \qquad ; \quad \chi''(\infty) = 0 \;\; \text{(finiteness)}$$

In the complex frequency plane, consider:

$$\psi(z) = \frac{\chi(z) - \chi(\omega)}{z - \omega}$$

Take the following contour integral:



$$\int_{-\infty}^{\omega-\epsilon} \frac{\chi(\omega') - \chi'(\infty)}{\omega' - \omega}\, d\omega' + \int_{\omega+\epsilon}^{\infty} \frac{\chi(\omega') - \chi'(\infty)}{\omega' - \omega}\, d\omega'$$

$$+ \pi i \left\{ \chi(\omega) - \chi'(\infty) \right\} = 0$$

Taking the real and imaginary parts:

$$\chi'(\omega) - \chi'(\infty) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\chi''(\omega')}{\omega' - \omega}\, d\omega'$$

$$\chi''(\omega) = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\chi'(\omega') - \chi'(\infty)}{\omega' - \omega}\, d\omega'$$

gives the KK relations.

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Recall:

$$\chi' - i\chi'' = \frac{\left(\frac{1}{2}\right)^{-1} \mathcal{N} \, |M_{ab}|^2}{\hbar \left(-\omega + \omega_{ba} + i T_2^{-1}\right)} \left(\rho_{aa} - \rho_{bb}\right)$$

$$i\hbar \dot{\rho}_{aa} = \mathcal{H}_{ab} \rho_{ba} - \rho_{ab} \mathcal{H}_{ba} - i\hbar \underbrace{T_1^{-1}}_{\text{random interactions with the reservoir}} \left(\rho_{aa} - \rho^{\circ}_{aa}\right)$$

with a similar equation for ρ_{bb}. This is for a two-level system. Now use the previous equations of motion for the off-diagonal elements.

$$i\hbar \dot{\rho}_{ab} = \mathcal{H}_{ab} \left(\rho_{bb} - \rho_{aa}\right) - i\hbar T_2^{-1} \rho_{ab}$$

Then:

$$\dot{\rho}_{bb} - \dot{\rho}_{aa} = \frac{-4\hbar^{-2} |\mathcal{H}_{ab}|^2 T_2}{\left(-\omega + \omega_{ba}\right)^2 T_2^2 + 1} \left(\rho_{bb} - \rho_{aa}\right) - \frac{\left(\rho_{bb} - \rho_{aa}\right) - \left(\rho^{\circ}_{bb} - \rho^{\circ}_{aa}\right)}{T_1}$$

In the steady state:

$$\rho_{bb} - \rho_{aa} = \frac{1}{1 + \dfrac{4\hbar^{-2} |\mathcal{H}_{ab}|^2 T_1 T_2}{\left(-\omega + \omega_{ba}\right)^2 T_2^2 + 1}} \left(\rho^{\circ}_{bb} - \rho^{\circ}_{aa}\right)$$

Resubstitution gives:

$$\chi'' = \frac{\left(\frac{1}{2}\right)^{-1} \hbar^{-1} |M_{ba}|^2 T_2}{\left(-\omega + \omega_{ba}\right)^2 T_2^2 + 1 + 4\hbar^{-2} |\mathcal{H}_{ab}|^2 T_1 T_2} \mathcal{N} \left(\rho^{\circ}_{bb} - \rho^{\circ}_{aa}\right)$$

For the magnetic field problem:

$$\mathcal{H}_{ab} = -M_{x\,ab} H_x \; ; \quad H_x = H_1 \cos \omega t$$

$$M_{x\,ab} = \frac{1}{2} \gamma \hbar$$

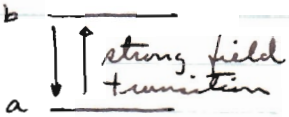
Then:

$$\chi'' = \frac{\frac{1}{2}\hbar\gamma^2 T_2}{1 + (-\omega + \omega_{ba})^2 T_2^2 + \frac{1}{4}\gamma^2 H_1 T_1 T_2} \; n\,(\rho^\circ_{bb} - \rho^\circ_{aa})$$

The above is for the linear case. Also, we have assumed that the relaxation process denoted by ρ is independent of the applied periodic field which is not generally true.

In the presence of a strong field, $\rho_{aa} - \rho_{bb} \to 0$.

Consider:



must greater than relaxation

$$W = \hbar^{-2} |\mathcal{H}_{ab}|^2 g(\nu)$$

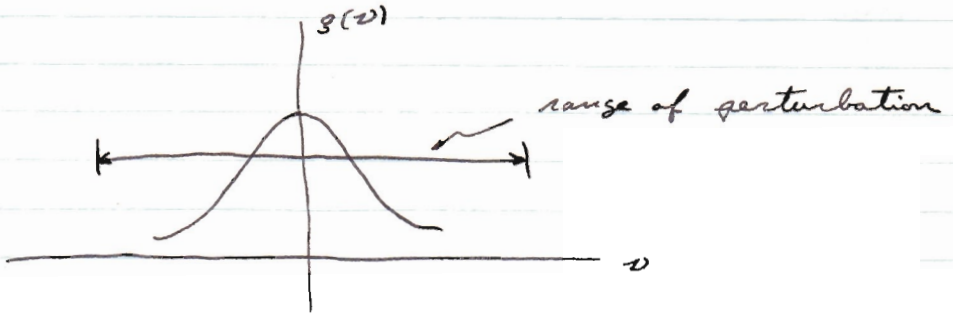
$$\frac{dN^b}{dt} = -W N_b + W N_a - w_{rel}\,(N_b - N_a - N_b^\circ + N_a^\circ)$$

with a similar expression for N_a. Then:

$$\frac{d(N_b - N_a)}{dt} = -2W(N_b - N_a) - \frac{1}{T_1}(N_b - N_a - N_b^\circ + N_a^\circ)$$

$$N_a - N_a = \frac{N_b^\circ - N_a^\circ}{1 + 2WT_1}$$ for the steady state saturation.

There is one situation where this form of the equation holds, and that is when the applied field is a random perturbation. That is:



This is the case for the optical maser.

The laser is pumped by a black body that is not uniform in angular distribution:

Cold: $kT \sim$ room temp (reservoir)

We know that the off-diagonal elements of ρ remain zero under a random perturbation, so that the population equations become those above. W becomes:

$$W_{BB} = \hbar^{-2} |\mathcal{H}_{ab}|^2 \underbrace{\varrho(\nu)}_{\text{density of BB oscillators}} = \frac{4\pi\nu^2}{c^3} d\Omega$$

Hence:

$$\frac{d(N_b - N_a)}{dt} = 2\frac{dN_b}{dt} = -\left(W_{pump}^{down} + W_{res}^{down}\right) N_b$$

$$+ \left(W_{pump}^{up} + W_{res}^{up}\right) N_a$$

Now: $\frac{W^{up}}{W^{down}} = e^{\frac{h\nu_{ba}}{kT}}$ so we can relate up and down

There is one other case in which a simple treatment is possible, viz, the strong Collision Model. Here, the free solution is applicable between collisions and at collision it returns to a thermal equilibrium situation.

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Strong Collision Model:

Strong collision at t_1:

$$\rho_{aa}(t_1) = \rho^{\circ}_{aa} \quad ; \quad \rho_{bb}(t_1) = \rho^{\circ}_{bb} \quad ; \quad \rho_{ab}(t) = 0$$

For the free spin, we have derived:

$$\rho_{aa}(t) - \rho_{bb}(t) = (\rho^{\circ}_{aa} - \rho^{\circ}_{bb}) \left[1 - 2\rho_{ba}(t - t_1) \right]$$

where: $\rho_{ba}(t - t_1) = \sin^2 2\theta \, \sin^2 \frac{1}{2}\omega_{eff}(t - t_1)$

$$= \frac{\gamma^2 H_1^2}{(\omega - \omega_{ba})^2 + \gamma^2 H_1^2} \sin^2 \tfrac{1}{2}\sqrt{(\omega - \omega_{ba})^2 + \gamma^2 H_1^2}\,(t - t_1)$$

This is the solution at time t following a collision at time t_1. We must now average over the possible collision times t_1. We take the distribution to be given by:

Probability to collide in interval t_1 and $t_1 + dt_1 = \dfrac{dt_1}{\tau_c}$

where τ_c^{-1} = no. of collision/sec.

This must be multiplied by the probability to have no collisions between dt_1+t_1 and t which is $e^{-(t - t_1)/\tau_c}$. We now find the stationary value of the density matrix by averaging over this distribution: $(t' = t - t_1)$:

$$\rho_{aa} - \rho_{bb} = (\rho^{\circ}_{aa} - \rho^{\circ}_{bb}) \int_0^{\infty} \{1 - \rho_{ba}(t')\} \, e^{-\frac{t'}{\tau_c}} \frac{dt'}{\tau_c}$$

$$= (\rho^{\circ}_{aa} - \rho^{\circ}_{bb}) \int_0^{\infty} \left[1 - \frac{\gamma^2 H_1^2}{\omega_{eff}^2} (1 - \cos \omega_{eff} t') \right] e^{-\frac{t'}{\tau_c}} \frac{dt'}{\tau_c}$$

or:

$$\rho_{aa} - \rho_{bb} = (\rho^{\circ}_{aa} - \rho^{\circ}_{bb}) \frac{1 + (\omega - \omega_{eff})^2 \tau_c^2}{1 + (\omega - \omega_{eff})^2 \tau_c^2 + \gamma^2 H_1^2 \tau_c^2}$$

We see $T_1 = T_2 = \tau_c$.

This is the result of Bloch with $T_1 = T_2 = T_c$. Recall the result for the susceptibility:

$$\chi' = \frac{\frac{1}{2}\gamma^2 \hbar T_2 (\omega_{ba} - \omega) T_2}{1 + (\omega_{ba} - \omega)^2 T_2^2 + \gamma^2 H_1^2 T_1 T_2} (\rho^0_{aa} - \rho^0_{bb})$$

$$\chi'' = \frac{\frac{1}{2}\gamma^2 \hbar T_2}{1 + (\omega_{ba} - \omega)^2 T_2^2 + \gamma^2 H_1^2 T_1 T_2} (\rho^0_{aa} - \rho^0_{bb})$$

where for the strong collision model we have $T_1 = T_2 = T_c$. Now define the time T_2':

$$T_2' \equiv \frac{T_2}{(1 + \gamma^2 H_1^2 T_1 T_2)^{1/2}}$$

Then:

$$\chi'' = \frac{\frac{1}{2}\gamma^2 \hbar}{(1 + \gamma^2 H_1^2 T_1 T_2)^{1/2}} \frac{T_2'}{1 + (\omega_{ba} - \omega)^2 T_2'^2}$$

which is a Lorentzian with the characteristic width T_2'. One cannot use the KK relations as the response function depends on H_1 and is hence non-linear. It can be seen, however, that:

$$\chi' = (\text{KK transform of } \chi'') \frac{T_2}{T_2'}$$

Thus the dispersion does not saturate as fast as the absorption does. It happens that in gaseous masers we have a distribution of resonant frequencies (density broadening). This means that we must average the above results over the distribution of resonant frequencies.

Take:

$$\omega_0 = \overline{\omega_{ba}} \quad ; \quad \omega_0' = \omega_{ba}$$

and the distribution as: $h(\omega_0' - \omega_0)$

If $h(\omega' - \omega_0)$ is Lorentzian with width T_2^*, we get a Lorentzian with:

$$\frac{1}{T_2^{\text{total}}} = \frac{1}{T_2'} + \frac{1}{T_2^*}$$

centered around $\overline{\omega_{ba}}$. However, seldom is h Lorentzian, it is more likely to be Gaussian due to Doppler broadening. In this case:

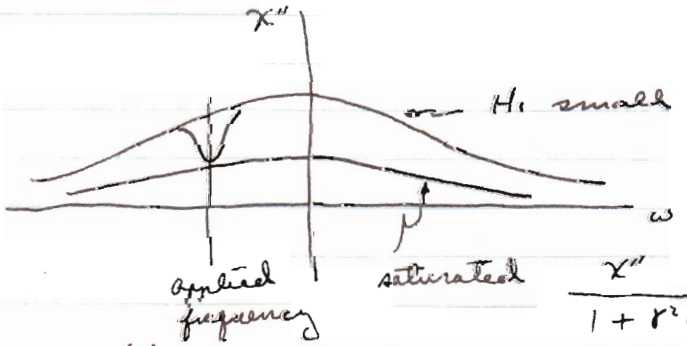
$$\omega' - \omega_0 = \Delta\omega_{\text{doppler}} \; ; \quad \frac{\Delta\omega_{\text{doppler}}}{\omega_0} = \frac{v_x}{c} \approx 10^{-5}$$

since v_x has a MB distribution, so does $\Delta\omega_{\text{doppler}}$:

$$h(\omega') = \frac{1}{\sqrt{2\pi \frac{kT\omega_0^2}{mc^2}}} \, e^{-\frac{(\omega' - \omega_0)^2 c^2 m}{2\omega_0^2 kT}} \, d\omega'$$

This effect usually determines the ultimate line-width of the light emitted by a gas. The rms width is: $\sqrt{\frac{kT\omega_0^2}{mc^2}}$. This width is very much larger than T_2'. Now, taking the Gaussian average over the strong collision model, one can consider the Lorentzian as a δ-function and hence the line shape is Gaussian, but remains saturated. The result is:

$$\chi'' = \frac{\frac{1}{4}\gamma^2 \hbar}{(1 + \gamma^2 H_1^2 T_1 T_2)^{1/2}} \; h(\omega - \omega_0)$$



(Burning a hole in a Gaussian distribution by driving it to saturation)

Note that χ' remains the same for both saturation and unsaturation. This is important for the operation of gas lasers. Also, the above treatment is good for electric dipole fields.

We have not treated homogeneous broading that occurs in a solid. Also inhomogeneous broadening occurs for changes in the ligand field throughout the solid.

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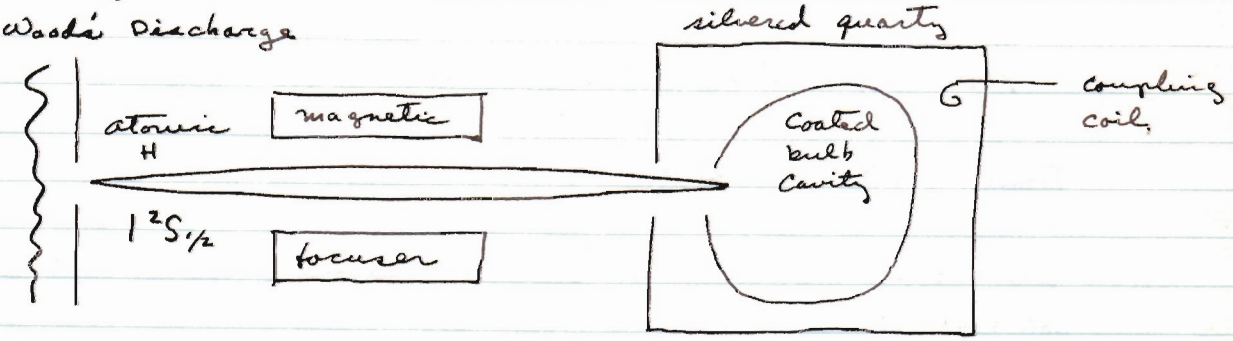
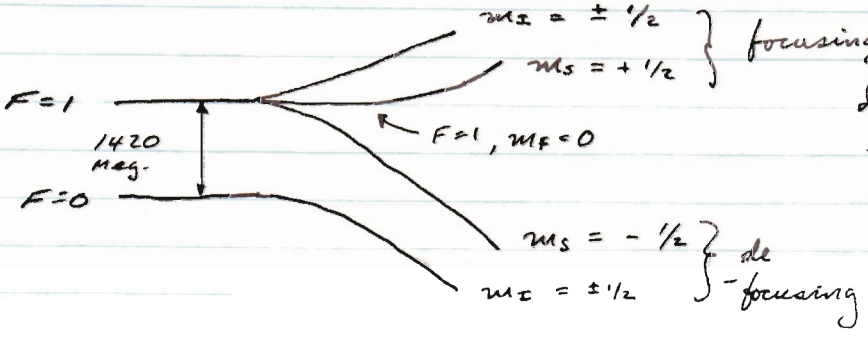
We now consider the problem of inverting the population of an atomic system. An inverted χ' was observed in 1931. Inverted populations have been observed since 1930, but it was Townes who realized the practical possibilities. His effort depended upon physical separation of the inverted atoms by means of passing a molecular beam through a magnetic field:

$$\underset{\sim}{F} = -\,\text{grad}\, W = \nabla(\underset{\sim}{M}\cdot\underset{\sim}{H}) = g\beta m_{II}\nabla|\underset{\sim}{H}|$$

This is the principle of the hydrogen beam maser (Ramsey).



The ground state of atomic H is 4-fold degenerate because of the proton spin. The levels are $F = I + S$:



It is seen that a separation of excited states can take place.

The separator-focuser coil has a cross-section like:

Hexapole Field

The magneto-static potential is derivable from Laplace's equation:

$$\frac{\partial^2 V_H}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial V_H}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 V_H}{\partial \theta^2} = 0$$

whose solution is:

$$V_H(r,\theta) = \sum_n \left(a_n r^n \cos n\theta + b_n r^n \sin n\theta\right)$$

Because of the symmetry, $a_n = 0$, and for r small, the leading term is b_3.

$$V_H = b_3 r^3 \sin 3\theta$$

$$H_r = -3 b_3 r^2 \sin 3\theta$$

$$H_\theta = -3 b_3 r^2 \cos 3\theta$$

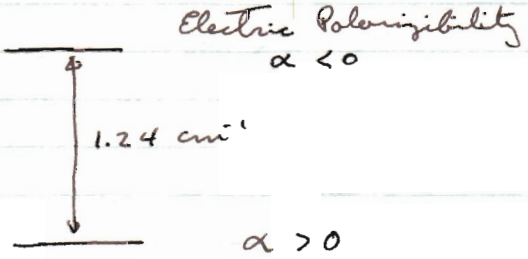
Then: $|H| = 3 b_3 r^2$

We see that there is a radial force on the particle:

$$F_3 = \pm \frac{1}{2} 3\beta \cdot 6 b_3 r$$

Thus we have a force proportional to the displacement providing focusing if we have the negative sign.

There is also the NH_3 molecular beam maser of Townes.



Thus we can use electric fields to effect separation, in which we use a focuser producing a quadrupole field:

$$V = a_2 r^2 \cos 2\theta$$
$$E_r = -2 a_2 r \cos 2\theta$$
$$E_\theta = -2 a_2 r \sin 2\theta$$
$$|E| = 2 a_2 r$$

Now: $\underset{\sim}{E} = -\text{grad } W = \nabla \frac{1}{2} \alpha |E|^2 = \alpha |E| \nabla |E|$

or:

$$F_r = 4 a_2^2 \alpha r \quad \text{(focusses for } \alpha < 0\text{)}$$

Hence we have a situation analogous to hydrogen.

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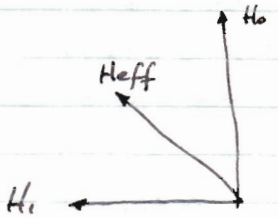
Adiabatic Rapid Passage:

Recall the treatment of this problem by Ehrenfest, that is, when the parameters of the problem are changed slowly with regard to the natural frequencies. In QM, this means that we do not induce transitions by the change. Consider:



Initially spin up. How fast can we change Heff and still have spin up? Require that:

$$\frac{1}{H_{eff}} \frac{dH_{eff}}{dt} << \gamma H_{eff}$$

The Fourier transform must not contain frequencies near the transition. Now:

$$\frac{dH_{eff}}{dt} = \frac{dH_0}{dt}$$

so that:

$$\frac{dH_0}{dt} << \gamma H_1^2 \leq \gamma H_{eff}^2$$

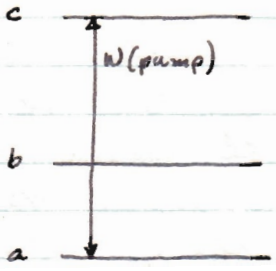
The "rapid" above means that the change must be short compared to T_2, T_1, so that the system can be considered free. This means: $\gamma H T_i >> 1$.

Instantaneous Reversal of External Field:

This means that H_0 changes so fast that the spin does not change so that the spins become aligned opposite the field and the system is then pumped up. A field of the order:

$H_0 \approx \frac{1}{\gamma T_2}$ has to be inverted in a time short compared to T_2. This only works in a magnetic spin system and in practice has only been done with nuclear spins.

Pumping With More Than Two Energy Levels:



We assume that the pumping radiation is non-coherent so that we can use the rate equations. We also assume the presence of some very weak, but coherent, signals. Call these: $S(\nu_{ca})$, $S(\nu_{ba})$.

Since the rate equations are still rigorous, we have:

$$\frac{dN_a}{dt} = W_{pump}\left(N_c - N_a e^{-\frac{h\nu_{ca}}{kT_{pump}}}\right) + W_{ca}\left(N_c - N_a e^{-\frac{h\nu_{ca}}{kT_{ca}}}\right)$$

$$+ W_{ba}\left(N_b - N_a e^{-\frac{h\nu_{ba}}{kT_{ba}}}\right) + \underbrace{S_{ba}}_{\frac{1}{4}\hbar^{-2}|M_{ba}|^2 H_i(\nu_{ba})\, g_{ba}(\nu)}\left(N_b - N_a\right)$$

$$\frac{dN_b}{dt} = W_{cb}\left(N_c - N_b e^{-\frac{h\nu_{cb}}{kT_{cb}}}\right) - W_{ba}\left(N_b - N_a e^{-\frac{h\nu_{ba}}{kT_{ba}}}\right)$$

$$- S_{ba}\left(N_b - N_a\right) + S_{cb}\left(N_c - N_b\right)$$

$$\frac{dN_c}{dt} = -\frac{dN_a}{dt} - \frac{dN_b}{dt}$$

Since we are interested in cw solutions, look at the steady state solution. Assume that the pump is very strong: $W_{pump} \gg \frac{\omega}{S}$. In fact, if $W \to \infty$, we see that $N_c = N_a$ or we have saturated these two levels. We have 3 possibilities in the steady state:

$N_b > N_c = N_a$: Stimulation at ν_{ba}

$N_b = N_c = N_a$: Heat Death

$N_b < N_c = N_a$: Stimulation at ν_{cb}

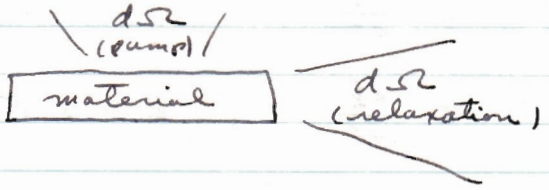
In the microwave case, we can achieve infinite pump frequency temperature. Although it is usual to have coherent pumps, we may easily imagine a non-coherent pump. We will assume a high temperature condition so we can linearize the exponentials. Then:

$$N_c - N_b = N_a - N_b = \frac{\omega_{ba}\gamma_{ba} - \omega_{cb}\gamma_{cb}}{\omega_{cb} + \omega_{ba} + S_{cb} + S_{ca}} \frac{h\nu}{kT_e}$$

where: $W_{pump} \sim \infty$

$T_e = T_{ca} = T_{cb} = T_{ba}$

Now consider the optical region. Relaxation takes place by spontaneous emission, because thermal effects are small in the optical region. The physical picture is:



Because all the fields involved are electromagnetic, we may regroup as follows:

$$\frac{dN_a}{dt} = A_{ca}N_c + B\rho(\nu_{ca})(N_c - N_a) \quad ; \quad B = \text{Einstein Coef.}$$
$$+ A_{ba}N_b + B\rho(\nu_{ba})(N_b - N_a)$$
$$+ S_{ba}(N_b - N_a)$$

where: $S_{ba} = \frac{1}{4}\hbar^{-2}|ex_{ba}|^2 E_s^2 g_{ba}$

or:

$$S_{ba} = A_{ba}\frac{c^2}{8\pi h\nu^3} \underbrace{I_s(\nu_{ba})}_{c\rho_s(\nu) = \frac{cE_s^2}{8\pi}} g_{ba}(\nu)$$

We have assumed here that the system is isotropic in taking $\rho(\nu)$ to be non-directional.

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Recall:

$$A_{ca} = \frac{64\pi^4 \nu_{ca}^3}{3hc^3} e^2 |x_{ca}|^2 \quad \text{(spontaneous)}$$

$$B = \frac{A c^3}{8\pi h \nu^3} \quad \text{(stimulated)}$$

$$f_{ca} = \frac{8\pi^2 m}{3h} \nu_{ca} |x_{ca}|^2 \quad \text{(oscillator strength)}$$

We now consider the coupling of a material system with one EM mode. Recall:

$$\underset{\sim}{E}(\underset{\sim}{r}, t) = -\sqrt{4\pi} \sum_\lambda p_\lambda(t) E_\lambda(\underset{\sim}{r})$$

as the normal mode expansion for the E-field. Now:

$$\nabla \times \underset{\sim}{E} = -\frac{1}{c} \frac{\partial \underset{\sim}{B}}{\partial t} \quad ; \quad \underset{\sim}{B} = \mu \underset{\sim}{H}$$

$$\nabla \times \underset{\sim}{H} = \frac{1}{c} \frac{\partial \underset{\sim}{D}}{\partial t} \quad ; \quad \underset{\sim}{D} = \epsilon \underset{\sim}{E} + 4\pi \underset{\sim}{P}_{ba}$$

$$\nabla \times \nabla \times \underset{\sim}{E} + \frac{\epsilon\mu}{c^2} \frac{\partial^2 \underset{\sim}{E}}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \underset{\sim}{P}_{ba}}{\partial t^2}$$

Note we have separated out the polarization due to one transition only, the rest being contained in ϵ. For normal modes:

$$\nabla \times \nabla \times \underset{\sim}{E}_\lambda(\underset{\sim}{r}) = k_\lambda^2 \underset{\sim}{E}_\lambda(\underset{\sim}{r})$$

$$\left[k_\lambda^2 p_\lambda(t) + \frac{\epsilon\mu}{c^2} \frac{\partial^2 p_\lambda}{\partial t^2} \right] (-\sqrt{4\pi}) \underset{\sim}{E}_\lambda(\underset{\sim}{r}) = 0$$

We have assumed that we have not taken out $\underset{\sim}{P}_{ba}$. We now see what the effect of this is. We also take ϵ complex and express it in terms of the cavity Q.

$$\left[\omega_\lambda^2 p_\lambda(t) + \frac{\omega_\lambda}{Q_\lambda} \dot{p}_\lambda(t) + \ddot{p}_\lambda(t) \right] (-\sqrt{4\pi}) \underset{\sim}{E}_\lambda(\underset{\sim}{r}) = -\frac{4\pi}{\epsilon\mu} \frac{\partial^2 \underset{\sim}{P}_{ba}}{\partial t^2}$$

Use: $\underset{\sim}{P}_{ba}(\underset{\sim}{r}) = \underset{\approx}{\chi}^{\omega}_{ba}\,(-\sqrt{4\pi})\,p_\lambda\,\underset{\sim}{E}_\lambda(\underset{\sim}{r})$

$p_\lambda = p_0\, e^{\imath\omega t}$ (assume eventually)

giving:

$$\left[\omega_\lambda^2\, p_\lambda(t) + \frac{\omega_\lambda}{Q_\lambda}\,\dot{p}_\lambda(t) + \ddot{p}_\lambda(t)\right](-\sqrt{4\pi})\,\underset{\sim}{E}_\lambda(\underset{\sim}{r})$$

$$= -\frac{4\pi}{\epsilon\mu}\,\underset{\approx}{\chi}_{ba}\,(-\sqrt{4\pi})\,\underset{\sim}{E}_\lambda(\underset{\sim}{r})\,\frac{\partial^2 p_\lambda(t)}{\partial t^2}$$

Operate with: $\int dV\ \underset{\sim}{E}_\lambda(\underset{\sim}{r})$

$$\left[\omega_\lambda^2\, p_\lambda(t) + \frac{\omega_\lambda}{Q_\lambda}\,\dot{p}_\lambda(t) + \ddot{p}_\lambda(t)\right] = -\frac{4\pi}{\epsilon\mu}\,\ddot{p}_\lambda(t)\,\underbrace{\frac{\int \underset{\sim}{E}_\lambda(\underset{\sim}{r})\cdot\underset{\approx}{\chi}_{ba}\cdot\underset{\sim}{E}_\lambda(\underset{\sim}{r})}{\int \underset{\sim}{E}_\lambda(\underset{\sim}{r})\cdot\underset{\sim}{E}_\lambda(\underset{\sim}{r})\,dV}}_{\eta\,\chi^{\omega}_{ba}}$$

η: filling factor

Now try the periodic solution:

$$\left[\omega_\lambda^2 - \omega^2 + \frac{\imath\omega_\lambda\omega}{Q_\lambda} - \frac{4\pi}{\epsilon\mu}\,\omega^2\,\eta\,\chi^{\omega}_{ba}\right] p_0 = 0$$

For a non-trivial solution, the $[\cdots]$ must $= 0$, giving two conditions:

Imaginary $= 0$: start Oscillation Condition:

$$\frac{\omega_\lambda\,\omega}{Q_\lambda} + \frac{4\pi}{\epsilon\mu}\,\omega^2\,\eta\,\chi''^{\omega}_{ba} = 0$$

(Can see that $\chi'' < 0$ is required)

Now: $\chi'' = \frac{1}{2}\,\hbar^{-1}\,|e x_{ba}|^2\,g_{ba}(\nu)\,(N_a - N_b)$

We then see, as a condition for oscillation:

$$N_b - N_a = \frac{\omega_d \in \mu}{\omega Q_d \hbar |e X_{ba}|^2 g_{ba}(\omega) \eta}$$

$N_b - N_a$ cannot exceed this value if oscillation is to be steady state. Unlike vacuum tubes, here we know something about the limiting non-linearity, which is the signal excitation itself (Recall: $\frac{}{\omega_{ba} + \omega_{ca} + S_{ca} + S_{ba}}$). Let us assume that we pump just above threshold, and assume $g(\omega)$ Lorentzian, giving for the saturated χ'':

$$\chi'' = \frac{\hbar^{-1} |e X_{ba}|^2 T_2 \left(N_a^{E_1=0} - N_b^{E_1=0}\right)}{1 + (\omega_b - \omega)^2 T_2^2 + \frac{1}{4} \hbar^{-2} |e X_{ba}|^2 E^2 T_1 T_2}$$

$$= \frac{\in \mu \omega_d}{\omega Q_d \eta}$$

This equation, thru E, gives exactly the level to which the oscillation builds up. We now find the frequency of oscillation from the real part of the eigenmode equation:

$$\omega_d^2 - \omega^2 - \frac{4\pi}{\in \mu} \omega^2 \eta \chi'_{ba}{}^{\omega} = 0$$

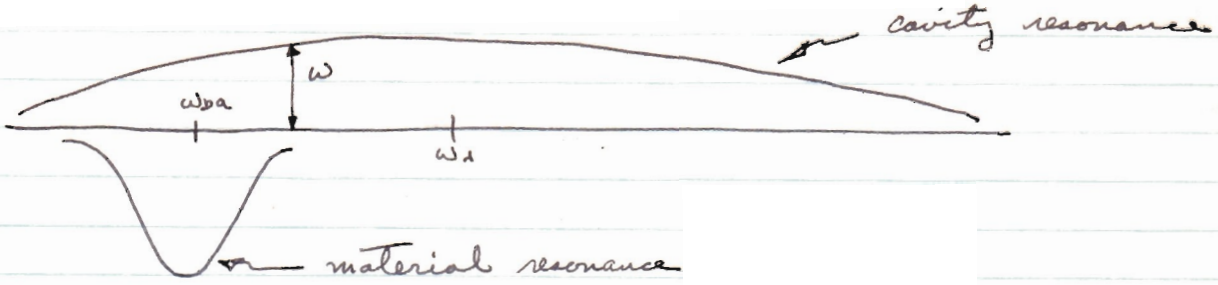
$$\omega^2 = \omega_d^2 + \frac{4\pi \omega^2 \eta \hbar^{-1} |e X_{ba}|^2 T_2 \left(N_a - N_b\right)_{E_1=0}}{\frac{\in \mu}{+ (-\omega + \omega_{ba})^2 T_2^2 + 1 + \frac{1}{4} \hbar^{-2} |e X_{ba}|^2 E^2 T_1 T_2}}$$

$$\cdot (\omega_{ba} - \omega) T_2$$

Substituting for E from χ'':

$$\omega^2 = \omega_d^2 + (\omega_{ba} - \omega) T_2 \omega_d \omega Q_d^{-1}$$

Graphical Picture:



Under the condition $\omega \approx \omega_d$; $\omega^2 - \omega_d^2 \sim 2\omega(\omega - \omega_d)$; and:

$$\omega - \omega_d = (\omega_{ba} - \omega)\frac{T_2 \omega_d}{2 Q_d}$$

Consider for cavity resonance: $\omega^2 - \omega_d^2 = 1 \omega_d \omega Q_d^{-1}$, suggesting:

$$\frac{T_2 \omega_d}{2 Q_d} = \frac{Q_d \text{ material}}{Q_d \text{ radiation}}$$

Then:

$$\omega = \frac{\omega_d Q_d + \omega_{ba} Q_{mat}}{Q_d + Q_{mat}}$$

From this we can see the intuitive results for $Q_d \gg Q_{mat}$ or $Q_d \ll Q_{mat}$. Both these conditions do occur in practice.

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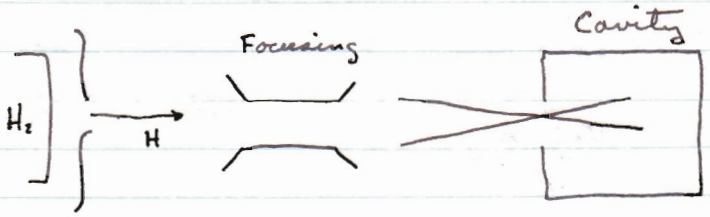
H_2 Beam Maser:



$F=1$ ——— $1/4\,A$

$F=0$ ——— $-3/4\,A$

Hyperfine interaction: $A\,\underset{\sim}{I}\cdot\underset{\sim}{S}$

$$\underset{\sim}{F} = \underset{\sim}{I} + \underset{\sim}{S}$$

$$\underset{\sim}{F}^2 = F(F+1) = I(I+1) + S(S+1) + 2\,\underset{\sim}{I}\cdot\underset{\sim}{S}$$

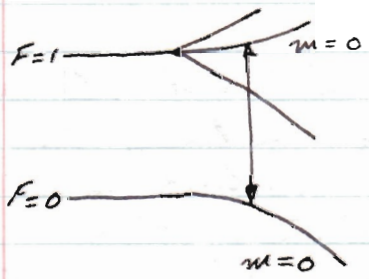
$$\underset{\sim}{I}\cdot\underset{\sim}{S} = \frac{1}{2}\left[F(F+1) - I(I+1) - S(S+1)\right]$$

Hence: $\langle \underset{\sim}{I}\cdot\underset{\sim}{S} \rangle_{F=1} = 1 - \frac{3}{4} = \frac{1}{4}$

$$\langle \underset{\sim}{I}\cdot\underset{\sim}{S} \rangle_{F=0} = 0 - \frac{3}{4} = -3/4$$

The total Hamiltonian is:

$$\mathcal{H} = A\,\underset{\sim}{I}\cdot\underset{\sim}{S} + g\beta\,\underset{\sim}{H}\cdot\underset{\sim}{S} + g_N\beta_N\,\underset{\sim}{H}\cdot\underset{\sim}{I}$$



Now: $A = h\nu_{hfs}$

where for H:

$\nu_{hfs} = 1,420,405,762.45 \pm .05$ cps

The cavity wall should be made of a magnetically inert material so that collisions with the wall do not change the H state. One of the best materials is Teflon. About 10^5 wall collisions can occur before a change of state.

The wall collision essentially determine the lifetime of the excited state. Also, particles can leak out thru the opening. Essentially:

$$\tau^{-1} = \tau_{esc}^{-1} + \tau_{\text{wall coll.}}^{-1} \quad ; \quad \tau_{esc} \sim 1/\text{sec}, \quad \tau_{\text{wall coll}} \sim 1/\text{sec}$$

where: $$\tau_{esc}^{-1} = \frac{\overline{|v_x|}\, A}{V}$$

Assume that $\mathcal{I}$ is the incident flux of H atoms, which is also the number escaping. Hence:

$$\mathcal{I} = (\text{shape factor})\ \frac{\overline{|v_x|}\, A}{V}\ N\, V$$

where N is the density. Solving for this:

$$N = \frac{\mathcal{I}\, \tau_{esc}}{V}$$

N has to exceed some critical value to cause oscillations.

The field inside the cavity is kept extremely low so that the hyperfine splitting is small:

$$\nu = h^{-1} \Delta E_{\substack{F=1,\, m=0 \\ F=0,\, m=0}} = h^{-1}\left(A + 2750\, H^2\right)$$

However, although the field in limited to a few milligauss, there will be variation in the microgauss range (kilocycles) which will cause transitions among the $F=1$ states, thus limiting the total lifetime by $\tau^{-1}_{\text{inhomo field}}$. This is a small quantity. Also we have spin exchange whose τ^{-1} is small. Thus we have a linewidth of the order of 1 cps.

Consider now the interaction matrix element:

$$\left\langle \frac{1}{\sqrt{2}}(\alpha_e \beta_N + \beta_e \alpha_N) \,\middle|\, g\beta S_z H_z \,\middle|\, \frac{1}{\sqrt{2}}(\alpha_e \beta_N - \beta_e \alpha_N) \right\rangle$$

$$= \frac{1}{2}\left[\frac{1}{2} g\beta + \frac{1}{2} g\beta\right] = \beta$$

We should include $g_N \beta_N I_z H_z$ but g_N is negligible so we neglect it.

We now have the necessary quantities to plug into our oscillation formulae. The "start oscillation" condition is:

$$\frac{\hbar^{-1} \beta^2 \tilde{T}_c \left(N_{\substack{F=1 \\ m=0}} - N_{\substack{F=0 \\ m=0}}\right)}{(-\omega + \omega_{HFS})^2 \tilde{T}_c^2 + 1 + \frac{1}{4}\hbar^{-2}\beta^2 H_{rf}^2 \tilde{T}_c^2} = \frac{\omega_c}{\omega Q 4\pi\eta}$$

This can be put into an energy balance form by multiplying them by $\frac{1}{2}\omega H_{rf}^2 V_{sample}$. Using $\frac{1}{Q} = \frac{1}{Q_c} + \frac{1}{Q_{wat}^{ex}}$, we have:

$$RHS = \frac{\omega_c W_c}{Q_c} + \frac{\omega_a W_c}{Q_{wat}^{ex}}$$

The LHS now is the power delivered by the atoms. From this we find the threshold flux to be:

$$\omega \hbar^{-1} \beta^2 \tilde{T}_c^2 \mathcal{A}_{th} = \frac{\omega_c W_c}{Q}$$

giving $\mathcal{A}_{th} \sim 10^{17}$ atoms/sec. If we exceed the threshold, we have a self-limiting condition:

$$\frac{\mathcal{A}}{1 + \frac{1}{4}\hbar^{-2}\beta^2 H_{rf}^2 \tilde{T}_c^2} = \mathcal{A}_{th}$$

This theory holds for all signal strengths because it is based on a strong collision model.

The power output is given by:

$$P_{out} = \frac{\omega_c W_c}{Q_{ex}} = \frac{\omega_c}{Q_{ex}} \frac{\hbar^2 V_{sample}}{\beta^2 T_c^2 8\pi\eta} \left(\frac{Q}{Q_{TH}} - 1\right)$$

$\approx 10^{-11}$ watts

From: $\omega - \omega_c = (\omega_{hfs} - \omega) \frac{T_c \omega}{2Q}$, it is found that ω is stable to within $1:10^{12}$ during 3 hrs. This is also approximately the resetability. The fundamental limitation is thermal fluctuations due to noise which sets the ultimate limit at $1:10^{15}$.

The NH_3 maser follows the exact Rabi solution because the lifetime is strictly limited by $\frac{l}{v}$ where l is the dimension of the cavity. This requires a different procedure than the H maser to calculate the power output.

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Noise in Masers

Circuit representation of the material system. Oscillation occurs when circuit impedance is zero and when zero phase shift occurs around the circuit. Define the material Q as:

$$Q_M = \frac{\omega \text{ (Energy stored)}}{\text{Power Absorbed by Material}}$$

This is negative for maser action. This Q_M should not be confused with the sharpness of the material linewidth. The linewidth is: $\Delta\nu_M = \frac{1}{2\pi T_c}$. $\nu/2\Delta\nu_M$ is the sharpness of the material, but is not in this case Q_M (but it could be defined as such). Our Q_M links the material with the circuit.

The oscillation condition requires that stimulated power equal absorbed power by cavity, or:

$$|Q_M| = Q \quad ; \quad Q_M < 0$$

where Q is the cavity Q.

Recall the transition probability:

Transition Probability Averaged Over Cavity

$$= \hbar^{-2} \beta^2 \overline{H_{rf}^2}\, g_M(\nu) \quad ; \quad g_M(\nu) = \frac{2T_c}{1 + 4\pi^2(\nu - \nu_M)^2 T_c^2}$$

and:

Power Absorbed at Material Resonance

$$= \hbar^{-1} \omega \beta^2 \overline{H_{rf}^2}\, 2T_c (N_a - N_b)$$

The energy stored is:

$$\text{Energy stored} = \frac{\int H_{rf}^2 \, dV}{8\pi} = \frac{\overline{H_{rf}^2} \, V_c}{8\pi} \qquad (= \bar{n} \, h\omega)$$

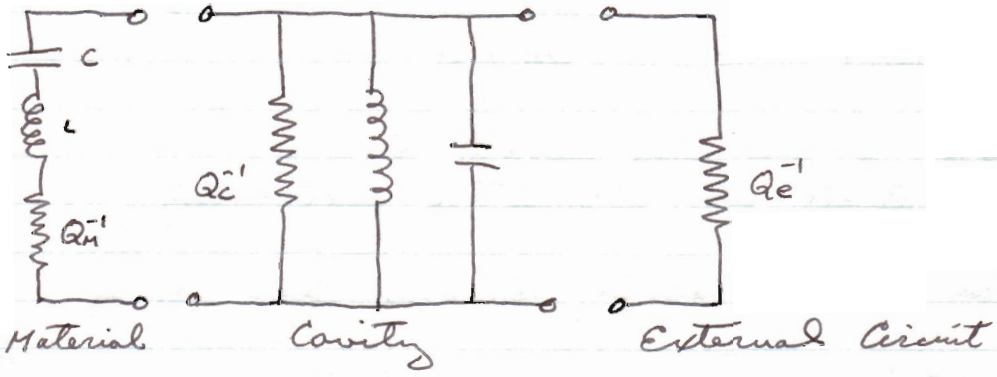
Hence:

$$Q_M^{-1} = \frac{8\pi \, h^{-1} \beta^2 \, z \, \tilde{\tau}_c}{V_c} (N_a - N_b)$$

We can say that the system has a conductance proportional to Q_M^{-1}.

Now we have reactive elements L and C which we choose to give oscillation and the proper material linewidth (Lorentzian). What if the linewidth is non-Lorentzian? Then we may use parallel L and C to approximate the true linewidth. Under inversion, L and C become negative (dispersive part of χ, while Q_M belongs to χ'').

The circuit is then:



We now have a representation independant of the material. However, we have not considered spontaneous emission which is the source of noise. We can represent this by a noise generator associated with Q_M.

For each mode, we have; for the probability of spontaneous emission:

$$\frac{\text{spon.}}{1} \quad \frac{\text{stimulated}}{+ \; \bar{n}_{ph}}$$

There will be a difference whether or not the cavity or material resonance is wider, and this appears in the choice of $g(\nu)$. We take the cavity to be broader. Then:

Spontaneous Emission Probability of Atom in Upper state into a Cavity Mode

$$= A_{cav} = \frac{8\pi h^{-1} \beta^2 Q}{V_c}$$

Recall for free space:

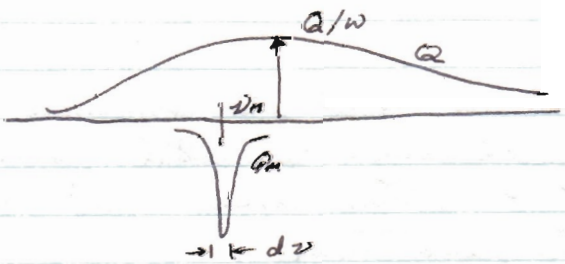
$$A_{free\ space} = \frac{64\pi^4}{h} \underbrace{\frac{\nu^3}{c^3}}_{\lambda^{-3}} \beta^2$$

$$\frac{A_{cav}}{A_{free\ space}} = \frac{\lambda^3}{V_c} \frac{Q}{4\pi^2}$$

For V_c in the order of the wavelength, we see that the spontaneous emission is enhanced by the cavity Q. This is because spontaneous emission that would usually occur at all wavelengths now occurs at the cavity wavelength. What is the noise power due to these spontaneous emissions in the cavity?

$$P_{noise} = h\omega N_b A_{cav}$$

We now ask for $P_{noise}(\Delta\nu)$; at material resonance:



(Assumed that $\nu_m \simeq \nu$)

$$P_{noise}(d\nu) = h\omega N_b A_{cav}\ 2\tilde{T}\ d\nu$$

Finally:

Noise Power Emitted in Tuned Cavity in Interval $d\nu$

$$= \frac{16\pi h^{-1}\beta^2 \omega \tau_c}{V_c} \frac{Q}{\omega} (N_a - N_b) \frac{N_b}{N_a - N_b} h\omega \, d\nu$$

$$= \frac{Q}{Q_M} \frac{h\nu}{e^{h\nu/kT_M} - 1} d\nu$$

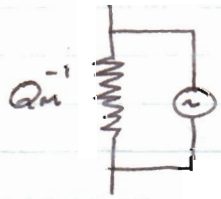
Circuit-wise, we are interested in the available noise power in a matched circuit ($Q/Q_M = 1$). Hence:

Noise Power Available from Material System (at $T_M > 0$)

$$= \frac{h\nu}{e^{h\nu/kT_M} - 1} d\nu \approx kT_M \, d\nu \quad \text{if } kT_M \gg h\nu$$

which is the usual Johnson Noise. We know we must have this because of very general thermodynamic arguments.

Thus we can postulate a noise generator equivalent circuit:



For inversion, it is hard to conceive of available noise power, whereas emitted power is still valid. Note that this does not change sign for negative temperatures.

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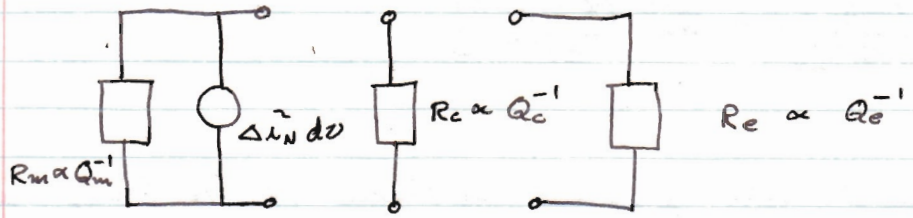
Recall the equivalent circuit (minus reactive elements):



where: $$\Delta i_N^2 \, d\nu = \frac{4 h\nu \, Q_M^{-1} \, d\nu}{e^{h\nu/kT_M} - 1}$$

This was derived under the assumption that the material resonance is sharper than the cavity resonance, and holds for negative temperatures. One can not show this by standard thermodynamic arguments. We have given a quantum (2 level) derivation of the Johnson noise, which we see arises from spontaneous emission and which is the source of all noise.

We get oscillation under the conditions: $Q_M < 0$; $Q_M^{-1} = Q_c^{-1} + Q_e^{-1}$. The oscillation amplitude is governed by the non-linearity of the material which we have determined analytically. It will be difficult, however, to account for the noise in a non-linear system. We will give an "ad hoc" argument. Consider first the system as an amplifier: $Q_M < 0$; $Q_c^{-1} < |Q_M^{-1}| < Q_c^{-1} + Q_e^{-1}$. We get for the power gain:

$$G_{max} = \left\{ \frac{Q_c^{-1} + Q_M^{-1} - Q_e^{-1}}{Q_c^{-1} + Q_M^{-1} + Q_e^{-1}} \right\}^2$$

where we are at resonance and Q_M is negative. For off-resonance, we must use:

$$G + jB = Q_M^{-1} \left\{ 1 + A \frac{(\nu - \nu_M)^2}{\Delta\nu_M^2} \right\}$$

For high gain; as a function of ν:

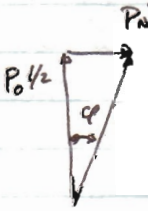
$$G(\nu) = \frac{(2Q_c^{-1})^2}{\left[\frac{(2Q_c^{-1})}{G_{max}^{1/2}} + Q_m^{-1}\frac{(\nu-\nu_m)^2}{\Delta\nu_m^2}\right]^2}$$

The gain-bandwidth product is taken at $1/2\ G_{max}$ for which:

$$\nu - \nu_m = \Delta\nu_m\ G_{max}^{-1/2}\ \frac{2Q_c^{-1}}{Q_c^{-1} + Q_c^{-1}}$$

Gaseous masers are narrow-band devices and hence are ~~good~~ bad amplifiers but good oscillators. In gaseous masers, the noise is generated mostly by the cavity walls. Here $T_m \approx -\infty$, so that the noise power from the material is $4kT|Q_m|^{-1}d\nu$ which is much less than the noise (at microwave frequencies) due to the cavity walls which is $4kTQ_c^{-1}d\nu$.

The out of phase component of the noise is more important than the in-phase component because this leads to a phase shift of the signal:



$$\langle\varphi^2\rangle = \frac{P_N^{\text{out-of-phase}}}{P_{out}} \quad \text{persist during } \tau_c$$

This leads to a random walk problem: The random walk of the phase is $\frac{t}{\tau_c}$ steps in time t:

$$\langle\Delta\varphi_t^2\rangle = \frac{\frac{1}{2}kT\,\Delta\nu_m}{P_{out}}\frac{t}{\tau_c} \approx 2\pi\tau_c^{-1} \quad ; \quad \tau_c^{-1} = 2\pi\Delta\nu_m$$

$$\langle\Delta\omega_t^2\rangle = \frac{kT}{P_{out}\,t}\frac{1}{\tau_c^2} = \frac{4\pi^2 kT}{P_{out}\,t}(\Delta\nu_m)^2$$

The observation time is the inverse of the frequency for a free-running oscillator.

$$\langle \Delta\nu^2 \rangle_{rms} = 2\pi \frac{kT}{P_{out}} \Delta\nu_m^2$$

Usually one sees a 2 instead of a 4, but this is unresolved as yet.

The point is that the frequency resolution for a H maser using the above formula is very good:

$$\frac{\Delta\nu_{rms}}{\nu_m} \approx 10^{16}$$

Consider for a moment a gaseous laser. Here $\lambda_m \sim 10{,}000\ \mathring{A}$, and the cavity is an FB interferometer. This cavity has a tremendous Q. The FB condition is:

$l = m\frac{\lambda}{2}$; $\nu_m = m\frac{c}{2l}$ where m is typically $2\cdot 10^6$. The Q of a mode, for mirrors with reflectivity R, is:

$$\frac{1}{Q_{opt}} = \frac{c\frac{E^2}{4\pi}(1-R)}{\omega\frac{E^2}{8\pi}l} = \frac{\lambda(1-R)}{\pi l}$$

For $m = 2\cdot 10^6$, $Q_{opt} = 10^8$. Now the sharpness of the material resonance is 10^5 so that the previous situation is now reversed. However, the modes are very close together:

$$\Delta\nu_m = \frac{c}{2l} \sim 150\ Mc$$

and therefore many modes are contained within a material linewidth.

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Gaseous Laser:

Consider an FB cavity, mirrors with reflectivity R. The mode condition is:

$$\nu_m = m \frac{c}{2l}$$

Take $l = 100$ cm, giving $\Delta \nu_m = 150$ Mc.

$$\frac{1}{Q} = \frac{\lambda (1-R)}{\pi l} \approx 10^{-8} \quad \text{for} \quad \lambda = 10{,}000\ \text{Å}.$$

The decay time for the mode is:

$$t_{decay} = \frac{Q}{2\pi\nu} = \frac{l}{2c(1-R)}$$

We note that the Q of the cavity is much greater than that of the atomic resonance. For a gaseous laser, there are about 10^3 modes in a linewidth. We ignore off-axis modes because of walkoff.

The on-axis mode pattern of two parallel mirrors, finite in size, can be calculated by forcing the transverse distribution of the E-field to repeat itself on reflection.

By Kirchoff's Law:

$$v(x',y') = \gamma \frac{e^{-\alpha|k|}}{4\pi} \iint u(x,y) \frac{e^{-\alpha|k|r}}{r} (1+\cos\theta)\, dx\, dy$$

$$u(x,y) = \gamma' \frac{e^{-\alpha|k|}}{4\pi} \iint v(x',y') \frac{e^{-\alpha|k|r}}{r} (1+\cos\theta)\, dx'\, dy'$$

We put these equal, resulting in a double integral equation which is solved by computer iteration. One does actually find a normal mode pattern. This is applicable to any shape of end-mirrors.

Another approach is to find the analytic solutions of Maxwell's equations:

$$\nabla^2 E_x - \frac{1}{c^2}\frac{\partial^2 E_x}{\partial t^2} = 0$$

Assume at constant z: $\nabla^2_{x,y} E_x = \frac{E_x}{\Lambda^2}$

This leads to Hermite functions. This is then put in the original equation.
See Gordon & Yariv, IEEE, Jan. '63.

We now discuss the start-oscillation condition by using Barkhausen's rule. That is, we put:

$$E_x^0 = E_x^0 r_1 r_2 e^{i/k/2l} \; ; \quad r_1 r_2 e^{i/k/2l} = 1 \; ; \; k = \epsilon'^{1/2}\frac{\omega}{c};$$

$$\epsilon = \epsilon_0 + 4\pi \chi_{ba}.$$

We linearize by putting $\epsilon'^{1/2} \approx 1 + 2\pi\chi'_{ba} \pm 2\pi i \chi''_{ba}$. If we assume the closed-loop gain to be small, we make an expansion of the exponential (this would not hold if the mirrors were poor reflectors). Also:

$$\mathrm{Re}\, r_1 r_2 = R = 1 - 4\pi \chi''_{ba}\, l \frac{\omega}{c}$$

$$e^{i/k/2l} \approx 1 + i/k/2l = 1 + 4\pi i \chi' l \frac{\omega}{c} - 4\pi \chi'' l \frac{\omega}{c}$$

Hence, the start oscillation condition is:

$$1 - R = -4\pi \chi''_{ba}\, l \frac{\omega}{c}$$

which we see as equal to our previous condition.

What is this in terms of population? Use the golden rule:

$$\tfrac{1}{2}\omega \chi''_{ba} |E|^2 = \hbar\omega (N_a - N_b) \frac{\hbar^2}{4} |\langle x_{ba}|^2 |E|^2 g_{ba}(\nu)$$

Hence, the critical inversion is:

$$N_b - N_a = \frac{(1-R)\hbar}{2\pi |\langle x_{ba}|^2 \ell \frac{\omega}{c} g_{ba}(\nu)} \approx 10^7/cc$$

Since the lifetime is 10^{-7} sec, this means we must pump 10^{14}/sec·cc. We see that lower frequencies and heavier atoms are better to use.

Which mode goes first? The one closest to the peak of $g_{ba}(\nu)$. In this sense, one should not excite other mode by pumping harder, but experimentally one observes this because of inhomogeneous saturation which leads to inhomogeneous broadening. One detects this by a photoelectric detector because this forms the square of the E-field which creates beats between the excited modes.

The doppler $g_{ba}(\nu)$ is:

$$g_{ba}(\nu) = \left(\frac{\hbar T}{2\pi M}\right)^{-1/2} \frac{c}{\omega} e^{-\frac{(\nu-\nu_m)^2 c^2 M}{2\hbar T \nu^2}}$$

The doppler width is half the gaussian:

$$\Delta\nu_D = 2\nu \sqrt{\frac{2\hbar T}{Mc^2} \ln 2}$$

Hence we can express the absorption coefficient in the familiar form:

$$\alpha = -\frac{1}{Q}\frac{dQ}{dz} = 4\pi\chi''\frac{\omega}{c} = \sqrt{\frac{\ln 2}{\pi}} \frac{A_{ba}}{4\pi} \frac{\lambda_D^2}{\Delta\nu_D} \left[-\frac{(N_b - N_a)}{g_b \; g_a}\right]$$

Sometimes the sign is reversed for ordinary absorption.

The frequency instability due to spontaneous emission is:

$$\delta\nu = \frac{4\pi h\nu}{P_{osc}} (\Delta\nu_c)^2$$

where $\nu = 3 \cdot 10^{14}$

$P = 1$ mwatt

$\Delta\nu_c = 1$ Mc

giving: $\delta\nu = 6 \cdot 10^{-3}$ cps

which is 2 parts in 10^{17}. This is not experimentally impossible because this would require the FB cavity dimension to remain constant to within a nuclear diameter.

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Originally, gaseous lasers were made with the FB mirrors inside the evacuated chamber, adjusted from the outside thru bellows. Now one uses Brewster windows which transmit 100% of the light polarized in the plane of incidence ($\tan \vartheta = n$, ϑ = angle of incidence).

In the He-Ne laser, the easiest excited line is $\nu_{osc} = 3 \cdot 10^{14}$ cps $= m \frac{c}{2l}$. This can be kept to within 1 part in 10^{12} for a few minutes (this was done at MIT in a wine cellar of a house on Cape Cod). The stability is checked by beating two lasers together since a photocell responds $\propto E^2$ giving an AC cross-term $\sim 2EE' \cos(\nu - \nu')t$.

Usually the FB mirrors are placed outside (with Brewster windows) on an Invar rod which can be tuned by magnetostriction. By this method, one can even obtain $\nu - \nu'$ in the audio range. $\nu - \nu' \sim 6$ KC was obtained by MIT for a few minutes.

We see that this laser is limited by external physical parameters rather than material noise. This laser could be used for a length standard rather than a time or frequency standard. The gaseous laser can measure lengths to one part in 10^{10}.

Now the Invar rod is subject to normal modes due to the presence of a finite temperature. This results in an accuracy of $1 : 10^{14}$ or a limit of $10^{-4} - 10^{-5}$ Å ($\frac{1}{2} kT \propto Y \cdot A (\Delta l)^2$). This effect could be eliminated by cooling.

What if the oscillator is off the cavity resonance? Recall:

$$\omega_{osc} - \omega_c = 2\pi \omega_{osc} \eta \chi'(\omega) \quad \text{(Inhomogeneous Broadening)}$$

where:

$$\chi'(\omega) = \frac{1}{4}\hbar^{-2}\,|e x_{ba}|^2\; Y(\omega)\; (N_b^\circ - N_a^\circ)$$

$Y(\omega)$ = KK transform of Gaussian

$$\chi''(\omega) = \frac{1}{4}\hbar^{-2}\,|e x_{ba}|^2\; h_{doppler}\; \frac{N_b^\circ - N_a^\circ}{\left\{1 + \frac{1}{4}\hbar^{-2}|e x_{ba}|^2 E^2 \gamma_c^{-2}\right\}^{1/2}}$$

Note $\chi'(\omega)$ does not saturate, so that we set a power dependent "pulling" effect due to $N_b^\circ - N_a^\circ$ being increased. The difference between adjacent modes due to this effect is:

$$\Delta\nu = \frac{c}{2\ell}\left\{1 - 2\pi\omega\,\frac{\partial\chi'(\nu)}{\partial\nu}\right\}$$

There is no pulling on the center mode because $\chi'(\nu) = 0$ but there is on adjacent modes.

Thus, for a length standard, one must keep the pumping power just above threshold.

The center frequency eats a hole in the doppler distribution, but the other modes eat two holes. One notes a drop in the power level when one tunes exactly to resonance.

Now, in the He-Ne laser, the upper level has $J=1$ and the lower $J=2$, so that magnetic field tuning is possible.

We now consider pumping mechanisms for gaseous lasers. Consider the rate equation for some level: (ground state denoted by o):

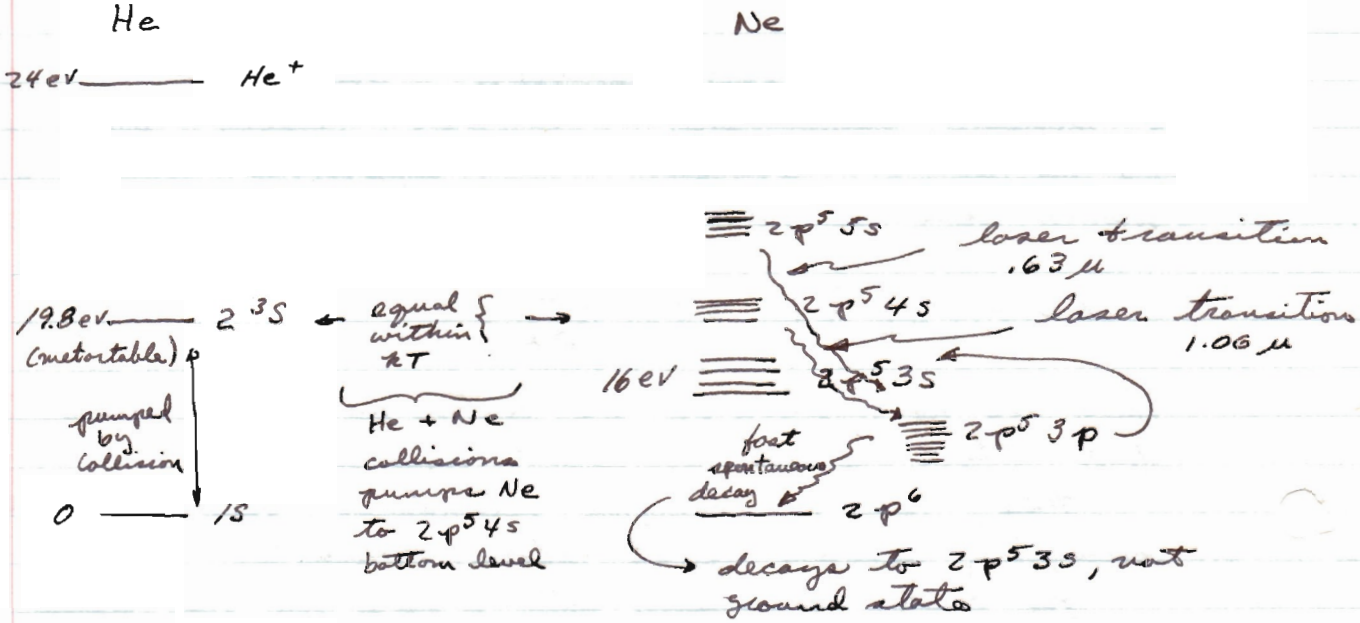
$$\frac{dN_k}{dt} = -\gamma_{diff} N_k - f_k A_{ko} N_k - \sum_{n}^{E_n < E_k} A_{kn} N_k - \underbrace{\sum_{n' \neq n_k} n'_j \bar{v}_{ij} Q_{oj} N_k}_{\text{inelastic collisions}}$$

$$+ B\,\rho(\nu_{ko})\,N_o + \sum_{n}^{E_n > E_k} A_{nk} N_n + \sum n_j \bar{v}_{jo} Q_{jo} N_o$$

where f_k is the imprisonment factor which retards the decay of k by spontaneous emission. γ_{diff} governs collision with the walls and depends on the diameter of the tube.

the third term is optical pumping, the fourth is spontaneous emission from higher levels and the fifth is pumping by inelastic collisions.

We now discuss pumping the He-Ne laser first discovered by Javan:



There are also some far infrared laser transitions, which have recently been discovered.

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Recall the effect of elastic modes on the FB cavity. The proper relation is:

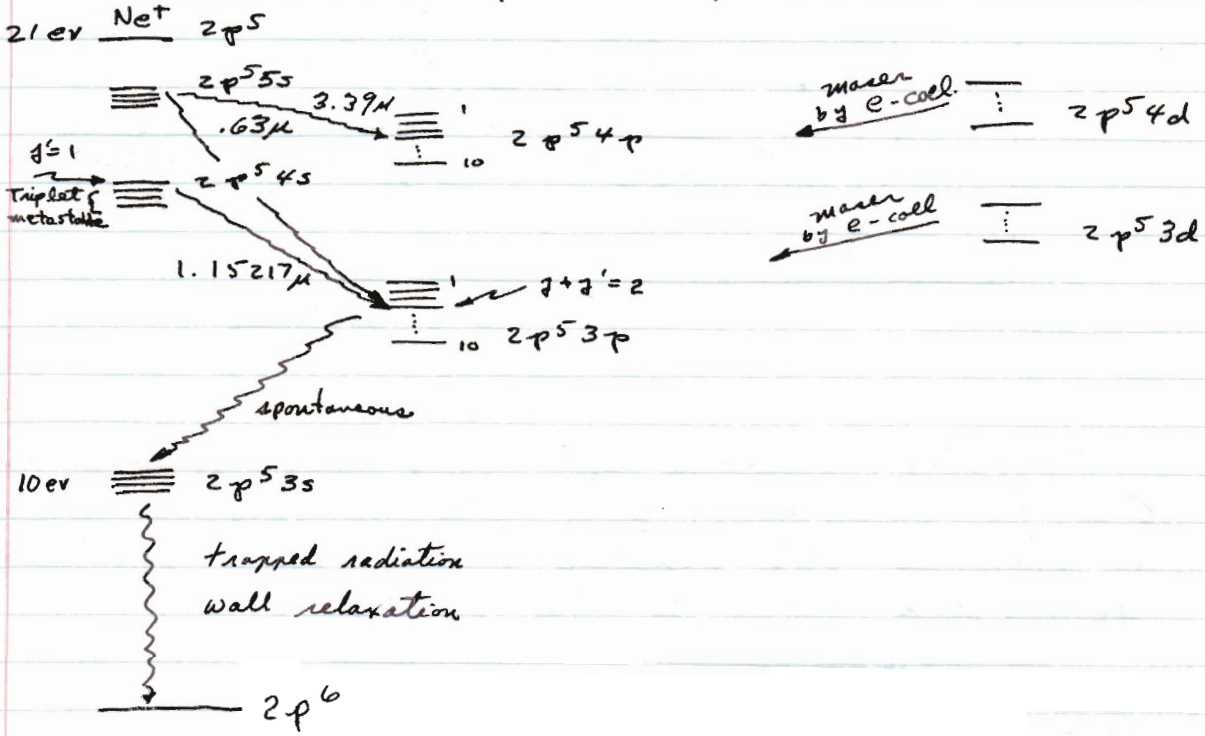
$$\tfrac{1}{2} kT = \tfrac{1}{2} Y_{\substack{\text{elastic} \\ \text{const.}}} \underset{\text{strain}}{e^2} V \; ; \qquad e^2 = \tfrac{1}{2} \frac{\overline{(\Delta l)^2}}{l^2}$$

The 1/2 comes from a spatial average over sinusoidal distribution of strain. Now:

$$\frac{\langle \Delta\nu \rangle_{rms}}{\nu} = \frac{\langle \Delta l \rangle_{rms}}{l} = \left(\frac{2kT}{YV} \right)^{1/2} \approx \begin{matrix} 1 \text{ part} \\ \text{in } 10^{14} \\ \text{at } 300^\circ K \end{matrix}$$

This effect can be minimized by cooling.

Consider the spectrum of the He-Ne mixture.



Note that 3.39μ and .63μ use the same upper level. The 3.39μ goes first because of narrower doppler broadening. One uses selective dielectric

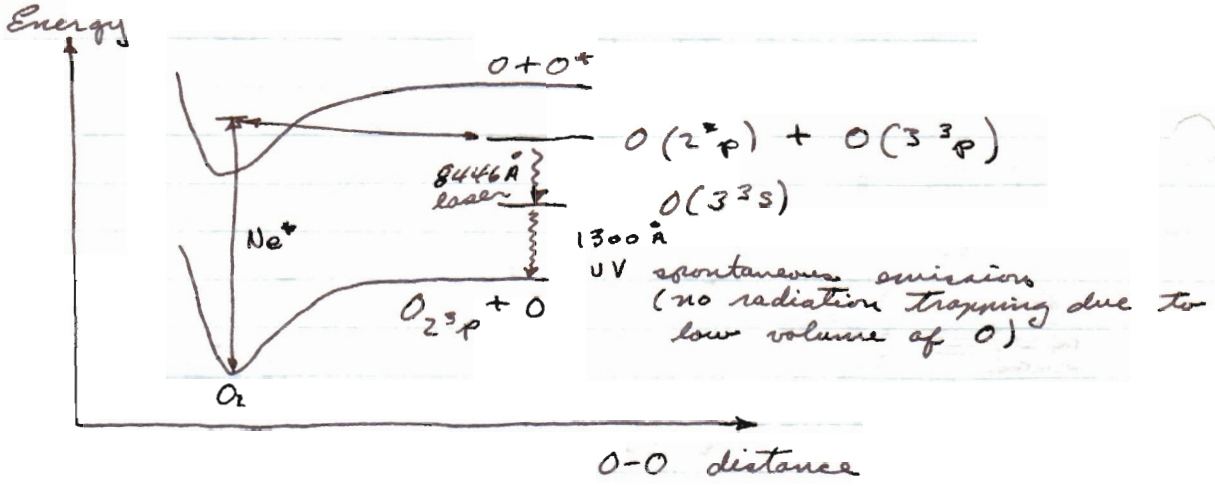
FB reflectors or a prism system to disperse unwanted radiation. Sharing of final states can create problems also.

One can excite by electron collisions the Ne directly without using He. These transitions are shown above. For all rare gases, there are now >100 different laser transitions, the longest wavelength being 28 μ.

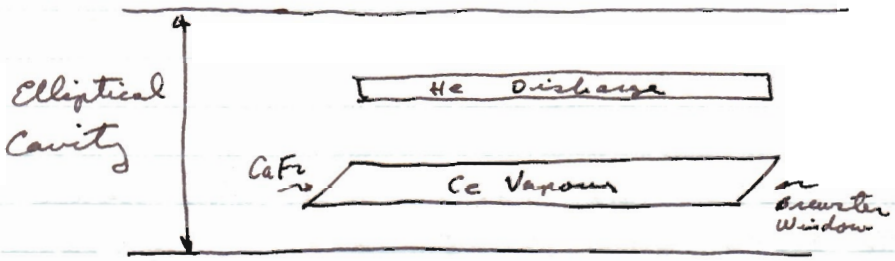
Another class of lasers are the dissociated excitation systems of Ne, A with O_2:

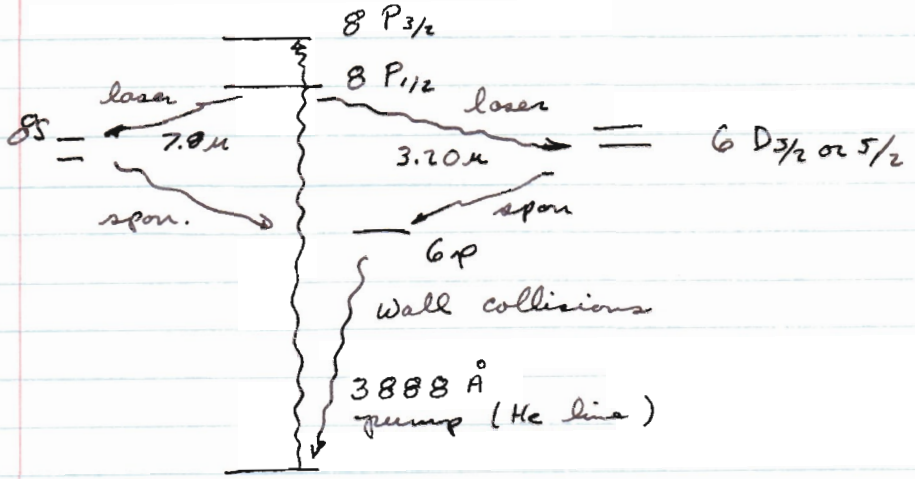
$Ne^* + O_2 \rightarrow O + O^* + Ne$ (* = metastable).

We excite the Ne triplet metastables.



We now consider optically pumped gaseous lasers. One has been made from Cesium vapour. This is because the doppler widths of a particular cesium line and a He line overlap.





This has been the only optically pumped gas laser.

Uses of gaseous lasers are:

(1) length standards

(2) IR spectroscopy sources (but power limited)

See Bennett's article on gaseous lasers.

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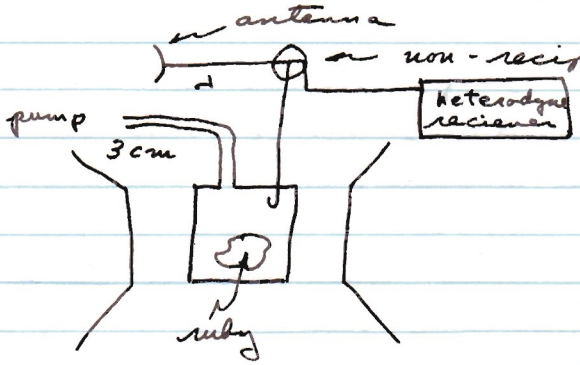
Reference Books: Singer, Masers
Sangyell, Lasers
Yarow & Gordon, Proc. IEEE, Jan '63.
Bennett } J. Appl. Optics
Herriott } Laser Supplement

Solid State Masers

For microwave operation, we need several well-defined levels in this wavelength region. The solids that have this are the paramagnetic solids where the spin transitions are very sharp. However, not spin 1/2 as this is only 2 levels. Also, for cw operation, we want a long T_1 or little coupling to lattice. To achieve this, one must cool. Note: $h\nu < kT$ down to 4.2°K. These solids obey Curie's law, very, population difference $\propto \frac{1}{T}$. Also, cavity Q goes up and the noise goes down according to the Johnson law. All this gives an effective noise temperature of the maser device of the order of a few degrees Kelvin. In 1956, this was about 1000 times better than anything available. Recently parametric devices have attained $T_N \sim 50°$. The range covered by masers is:

1 mm — 100 cm

or 300 KMC — 300 MC



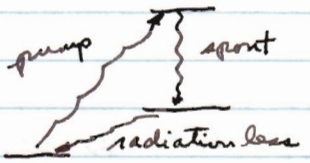
Cr^{3+} in Al_2O_3

Take ν_s = 1420 MC

By saturating the pump, the device can be made extremely stable in gain.

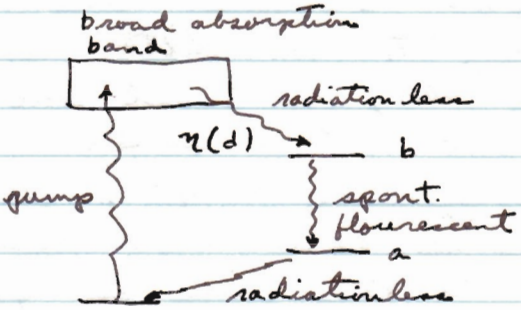
this maser can be only used on antennas that look up because of the high noise temperature of the earth's surface.

We now consider solid state Optical Masers. A necessary condition is the presence of flouresence in the solid. These lines must also be narrow. The ideal situation is:



This is not the case in ruby, where the ground state and the final state for radiation are the same.

A more typical situation is (4 level system):



The paramagnetic solids meet these requirements in most respects. $\eta(d)$ is the flourescent efficiency.

The solid should have good optical properties and this is usually the limiting factor.

The threshold condition is:

$$\int \frac{\eta(d)\, I(d)\, d\kappa(d)}{hc}\, dd = (N_b^{th} - N_a^{th})\, A_{ba}$$

where $I(d)$ is the pump power, $\kappa(d)$ the absorption coefficient. A typical value for the incident light power is 20 watts/cm^2.

We now consider some of the Quantum mechanics of paramagnetic salts:

$$\mathcal{H} = \sum \frac{1}{2m}\left(\underset{\sim}{p} - \frac{e}{c}\underset{\sim}{A_0}\right)^2 + V_{ion} + V_{cr} + \sum \lambda \underset{\sim}{l}\cdot\underset{\sim}{s}$$

$$+ \sum_{\text{spin - other orbits}} + \mathcal{H}_{HFS} + g\beta\, \underset{\sim}{S}\cdot\nabla\times\underset{\sim}{A_0}$$

Take RS coupling and $\underset{\sim}{A_0} = \frac{1}{2}\underset{\sim}{H_0}\times\underset{\sim}{r}$:

$$\mathcal{H} = \underbrace{\sum \frac{p^2}{2m} - \sum \frac{Ze^2}{r} + \sum\sum \frac{e^2}{r}}_{10^5\ cm^{-1}} + \underbrace{\lambda\, \underset{\sim}{L}\cdot\underset{\sim}{S}}_{10-10^{+3}\ cm^{-1}}$$

$$+ \underbrace{\beta\left(\underset{\sim}{L} + 2\underset{\sim}{S}\right)\cdot\underset{\sim}{H_0}}_{1\ cm^{-1}\ \text{for}\ 10\ kg} + \underbrace{\mathcal{H}_{HFS}}_{10^{-2}-10^{-1}\ cm^{-1}} + \underbrace{\mathcal{H}_{diam}}_{10^{-4}\ cm^{-1}\ \text{in}\ 10\ kg\ \text{field}}$$

$$+ \underbrace{V_{cryst.}^{\text{cubic}}}_{10^2-10^4\ cm^{-1}} + \underbrace{V_{cryst}^{\text{other sym}}}_{1-10^2\ cm^{-1}}$$

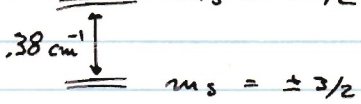
Further developments depend on the order of the application of perturbation theory. In the iron group, one should take $V_{cryst.}$ before $\lambda\, \underset{\sim}{L}\cdot\underset{\sim}{S}$. See Griffith. At any rate, this $\mathcal{H}$ gives both microwave and optical spectra.

Consider Cr^{3+} in Al_2O_3, trigonal symmetry with a strong cubic component. We begin with 3 3d electrons. Hund's Rule gives an F state with $S = 3/2$. The SO coupling gives:

ground state ——— an spin quartet

$S = 3/2$

This level splits in V_{cryst} to



This is the microwave transition and can be tuned by an H-field parallel to the trigonal axis. The spin Hamiltonian is:

$$\mathcal{H}_{spin} = D\left\{S_z^2 - \frac{1}{3}S(S+1)\right\} + \beta\, \underset{\sim}{S}\cdot\underset{\sim}{g}\cdot\underset{\sim}{H} + A\,\underset{\sim}{I}\cdot\underset{\sim}{S} + \mathcal{H}_Q$$

$$+ g_z\beta_N\, \underset{\sim}{I}\cdot\underset{\sim}{H}$$

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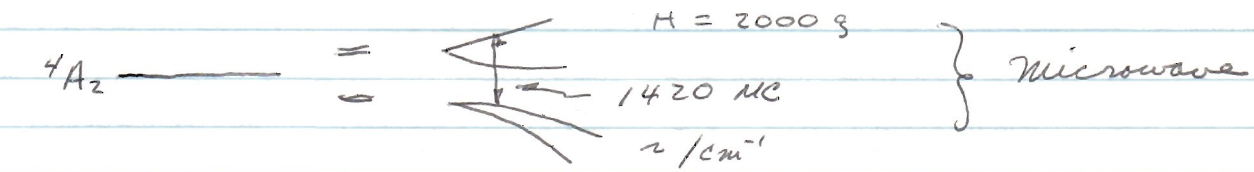
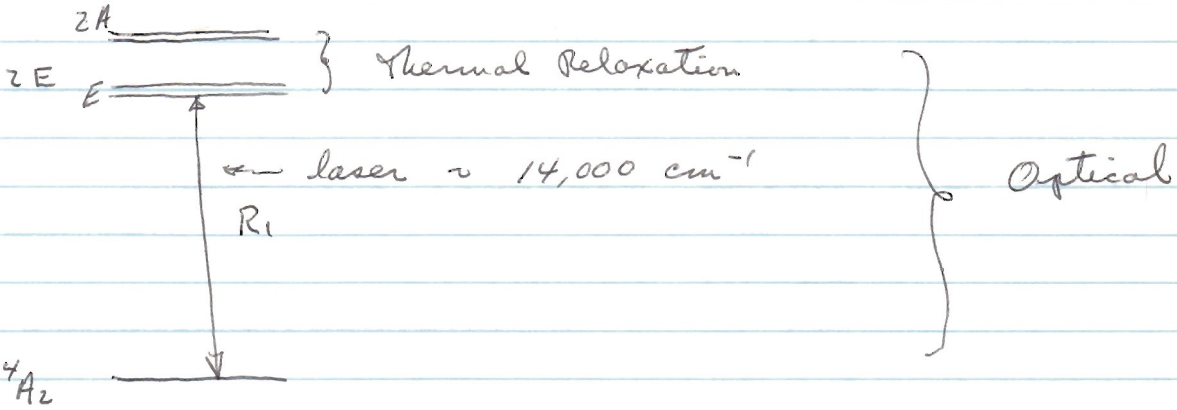
For details on traveling wave masers, see:
de Grasse, Scovil, BSTJ
Treacy, 3rd Conf. Q. El.
Jelly, IEEE Q. El. Issue

Recall the spin Hamiltonian. For further discussion, we assume that this H has been diagonalized. We consider the ruby states.



Absorption Band



We concentrate on the 1420 MC maser. The spin Hamiltonian is:

$$\mathcal{H}_{spin} = D\left[S_z^2 - \tfrac{1}{3}S(S+1)\right] + g\beta\, \underline{S}\cdot\underline{H}$$

whose solution gives:

$H_2 \rightarrow$

Although we have considered the ions independent, they actually interact there a magnetic dipole field. This leads to a broadening of the lines, sometimes washing them out completely for a high concentration of impurities. The form of the interaction is:

$$\mathcal{H}_{dip} = \sum_{i<j} \frac{h^{+2} \gamma_i \gamma_j \beta^2}{r_{ij}^3} \left[\underset{\sim}{S}_i \cdot \underset{\sim}{S}_j - \frac{3(\underset{\sim}{S}_i \cdot \underset{\sim}{r}_{ij})(\underset{\sim}{S}_j \cdot \underset{\sim}{r}_{ij})}{r_{ij}^2} \right]$$

Also we have an exchange interaction:

$$\mathcal{H}_{ex} = \sum_{i<j} A_{ij} \underset{\sim}{S}_i \cdot \underset{\sim}{S}_j$$

These interactions introduce the so-called adiabatic broadening which results not from lifetime considerations, but from a multiplicity of states.

Assume we know the 4 states of two neighboring ions:

d — d' —

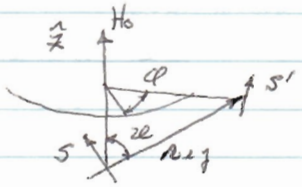
c — c' —

b — b' —

a — a' —

a diagonal element would be $(a, b' | \mathcal{H}_{dip} | a, b')$ where a semi-diagonal term would be $(a, b' | \mathcal{H}_{dip} | b, a')$. In general $\mathcal{H}_{dip}$ connects all states so there are many off-diagonal elements. If we consider $D = 0$, the problem is simplified.

$3/2$ — d — d'
$1/2$ — c — c'
$-1/2$ — b — b'
$-3/2$ — a — a'



Now: $$\mathcal{H}_{dip} = \frac{\gamma^2 \hbar^2}{r_{ij}^3} (A + B + C + D + E + F)$$

where:

$$A = S_z S_z' (1 - 3\cos^2\theta) \quad \{\text{diagonal}\}$$

$$B = -\frac{1}{4} (S^+ S^- + S^- S'^+) (1 - 3\cos^2\theta) \quad \{\text{semi-diagonal}\}$$

$$C = -3/2 (S^z S'^+ + S^+ S'^z) \sin\theta \cos\theta e^{-i\varphi}$$

$$D = C^* (S^z S'^- + S^- S'^z)$$

$$E = -\frac{3}{4} S^+ S'^+ \sin^2\theta e^{-2i\varphi}$$

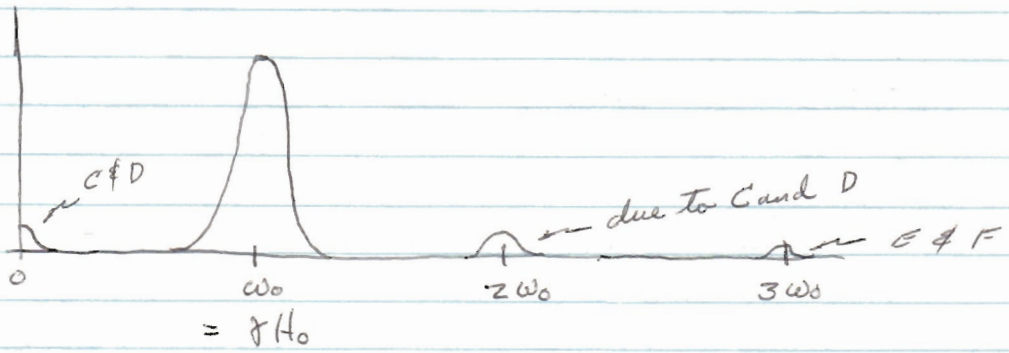
$$F = E^*$$

To proceed further, we must truncate the off-diagonal elements in order to see how broadening enters when an rf field is applied. The off-diagonal elements form states which are admixtures of the former states. If:

$$\frac{\frac{\gamma^2 \hbar^2}{r^3}}{\gamma \hbar H_0} \ll 1 \quad ; \text{ or } \quad \frac{H_{loc}}{H_0} \ll 1$$

Then truncation can be defined as the broadening is much less than the resonance.

Consider:



The broadening is then caused by the diagonal and ~~off~~ semi-diagonal terms. It is calculated by the method of moments.

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We now calculate the broadening by the method of moments (Waller, Van Vleck). Since the result is a trace, we may use the unperturbed representation in the calculation. The interaction $\mathcal{H}$ is:

$$\mathcal{H}_{rf} = M_x H^x_{rf}$$

We form the normalized second moment of the absorption curve:

$$\langle \omega^2 \rangle = \frac{\sum_n \sum_{n'} M_{xnn'} M_{xn'n} \omega^2_{nn'} \rho_{nn}}{\sum_n \sum_{n'} M_{xnn'} M_{xn'n} \rho_{nn}}$$

This may be carried out for all the moments thus defining an analytic function.

Now note:

$$\dot{M}_{xnn'} = i \omega_{nn'} M_{xnn'}$$
$$\dot{M}_{xn'n} = i \omega_{n'n} M_{xn'n}$$

from which we form:

$$\dot{M}_{xnn'} \dot{M}_{xn'n} = \omega^2_{nn'} M_{xnn'} M_{xn'n}$$

This would be similar to the $\langle \omega^2 \rangle$ if ρ_{nn} = constant. Such is the high T case, $kT \gg h\nu$ for which we have the equilibrium value of ρ_{nn}.

We then see:

$$\langle \omega^2 \rangle_{\substack{\text{high} \\ \text{Temp}}} = \frac{Tr\{\dot{M}_x^2\}}{Tr\{M_x^2\}}$$

Now: $i\hbar \dot{M}_x = [\mathcal{H}, M_x]$; $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{dip}$
and thus:

$$\langle \omega^2 \rangle_{HT} = \frac{-\hbar^{-2} \, Tr\{[\mathcal{H}, M_x]^2\}}{Tr\{M_x^2\}}$$

We note that the cross-term $Tr\, \mathcal{H}_0 \mathcal{H}_{dip} = 0$ because it is odd in the spin.
We have: $\mathcal{H}_0 = g\beta H_0 S_z$; $M_x = -g\beta S_x$, so that:

$$\langle \omega^2 \rangle_{HT} = \underbrace{\frac{-\hbar^{-2} \, Tr\{[\mathcal{H}_{dip}, M_x]^2\}}{Tr\{M_x^2\}}}_{\langle \Delta\omega^2 \rangle} + \underbrace{\omega_0^2}_{\text{can see this from physical grounds}}$$

Now, to get correct results, we must truncate the C, D, E, F terms of $\mathcal{H}_{dip}$ because they contribute via satellite humps very heavily to $\langle \Delta\omega^2 \rangle$, Because we are interested in the width of the main peak, we eliminate these terms. For unlike neighbors, we must also eliminate B as this gives a separate resonance peak.

Similarly, we have:

$$\langle \Delta\omega^4 \rangle = \frac{\hbar^{-4} \, Tr\{[\mathcal{H}_{dip}', [\mathcal{H}_{dip}', M_x]]^2\}}{Tr\, M_x^2}$$

In the case of an external non-crystal field (magnetic) we can evaluate the traces: Consider:

$$\gamma^2 \hbar \sum_{j<k}' \sum_k' r_{jk}^{-3} \left[A_{jk} + B_{jk}, \sum_l S_{xl} \right]$$

$$= \gamma^2 \hbar \sum_{j<k}' \sum_k' r_{jk}^{-3} \frac{3}{2} (1 - 3\cos^2 \vartheta_{jk}) \left[I_{zj} I_{zk}, I_{xj} + I_{xk} \right]$$

$(I \equiv S)$

Eventually:

$$Tr\left\{\left[\mathcal{H}_{dip}^{T}, \Sigma S_x\right]^2\right\} = \frac{1}{2}\gamma^4\hbar^2 S^2(S+1)^2(2S+1)^N \cdot \sum_{j<k}\sum_{k} r_{jk}^{-6}\left(1-3\cos^2\theta_{jk}\right)^2$$

$$Tr\left\{\left(\Sigma S_x\right)^2\right\} = \frac{1}{3} S(S+1)(2S+1)^N$$

Hence:

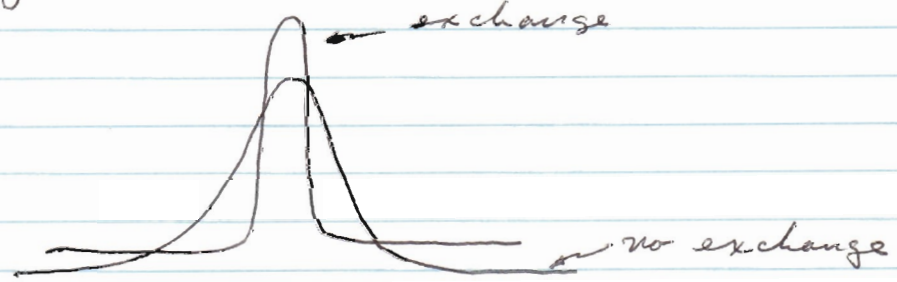
$$\langle \Delta\omega^2\rangle_{\substack{\text{dipolar}\\ \text{like spins}}} = \frac{3}{4}\gamma^4\hbar^2 S(S+1)\sum_{k}\frac{\left(1-3\cos^2\theta_{kj}\right)^2}{r_{kj}^6}$$

By calculating $\langle \Delta\omega^4\rangle$, we find that if we assume a gaussian line shape, it is a good approximation; ($\langle\Delta\omega^2\rangle^2 \approx 3\langle\Delta\omega^4\rangle$). For unlike spins (eliminate B) we get 1/3 instead of 3/4 in $\langle\Delta\omega^2\rangle$.

For an exchange interaction ($J_{kj}\,\underset{\sim}{S}_k\cdot\underset{\sim}{S}_j$), we see:

$$\left[\underset{\sim}{S}_k\cdot\underset{\sim}{S}_j,\ S_{xk}+S_{xj}\right] = 0 \quad \text{for like spins.}$$

This means that the exchange interaction does not contribute to $\langle\Delta\omega^2\rangle$ but it does to $\langle\Delta\omega^4\rangle$ because: $\left[\mathcal{H}_{ex}\left[\mathcal{H}_{dip}, S_x\right]\right] \neq 0$.
Physically this means:



Since we cannot observe tails, we have <u>exchange narrowing</u>. For unlike spins, we get <u>exchange broadening</u>. Both effects occur in masers.

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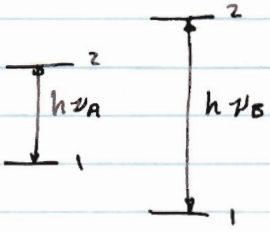
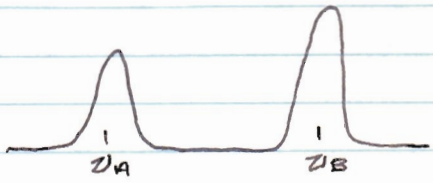
Consider two unlike spins:





$$W_{cross} = \hbar^{-2} \left| B_{AB}^{S_A^+ S_B^-} \right|^2 g_{cross}(\nu=0)$$

where:

$$g_{cross}(\nu = \nu' - \nu'') = \int g_A(\nu' - \nu_A)\, g(\nu'' - \nu_B)\, d\nu'\, d\nu''$$

This calculation is based on the "overlap" model.

Recall the rate equations: $\Delta n_A = n_{A1} - n_{A2}$

$$\frac{d(\Delta n_A)}{dt} = -W_{pump}\,\Delta n_A - \frac{1}{T_{1A}}\left(\Delta n_A - \frac{1}{2} N_A \frac{h\nu_A}{kT}\right)$$

To this must be added the cross terms:

$$- W_{cross}\, n_{A1} \frac{n_{B2}}{N_B} + W_{cross}\, n_{A2} \frac{n_{B1}}{N_B}$$

Similarly:

$$\frac{d(\Delta n_B)}{dt} = -\frac{1}{T_{1B}}\left(\Delta n_B - \frac{1}{2} N_B \frac{h\nu_B}{kT}\right) - W_{cross}\, n_{B1} \frac{n_{A2}}{N_A} + W_{cross}\, n_{B2} \frac{n_{A1}}{N_A}$$

We linearize by:

$$n_{A1} \to \tfrac{1}{2} N_A + \tfrac{1}{2} \Delta n_A \;; \quad n_{B2} \to \tfrac{1}{2} N_B - \tfrac{1}{2} \Delta n_B \;;$$

$$n_{A2} \to \tfrac{1}{2} N_A - \tfrac{1}{2} \Delta n_A \;; \quad n_{B1} \to \tfrac{1}{2} N_B + \tfrac{1}{2} \Delta n_B$$

This allows:

$$\frac{d(\Delta n_A)}{dt} = -W_{pump}\,\Delta n_A - \frac{1}{T_{1A}}\left(\Delta n_A - \tfrac{1}{2} N_A \frac{h\nu_A}{kT}\right) - W_{cross}\,\tfrac{1}{2}\left[\Delta n_A - \frac{N_A}{N_B}\,\Delta n_B\right]$$

$$\frac{d(\Delta n_B)}{dt} = -\frac{1}{T_{1B}}\left(\Delta n_B - \tfrac{1}{2} N_B \frac{h\nu_B}{kT}\right) - W_{cross}\,\tfrac{1}{2}\left(\Delta n_B - \frac{N_B}{N_A}\,\Delta n_A\right)$$

Now take $N_A = N_B$; $W_{pump} \to \infty$; $\Delta n_A \sim 0$; so that in the steady state:

$$\Delta n_B = \frac{\frac{1}{T_{1B}}\left(\frac{1}{2} N_B \frac{h\nu_B}{kT}\right)}{\frac{1}{T_{1B}} + \frac{1}{T_{cross}}}$$

If $T_{cross} < T_{1B}$:

$$\Delta n_B = \frac{T_{cross}}{T_1}\,(\Delta n_{eq})$$

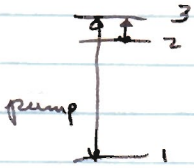
Now: $T_2 \sim 10^{-7} - 10^{-8}$ sec ; $T_1 \sim 10^{-1} - 10^{-3}$ sec @ 4°K.

The cross-relaxation process is not as simple as we have indicated. Generally:

$$\frac{dn_\alpha}{dt} = W_{pump_{ij}}(n_j - n_\alpha) - W_{ij}\left\{n_\alpha - n_j - n_i^\circ + n_j^\circ\right\} + W^{cross}_{ij\,k\ell}\,\frac{1}{N}\left(n_k n_\ell - n_\alpha n_j\right) + \underbrace{W^{cross}_{ij\,k\ell\,mnop}\,\frac{1}{N^3}\left(n_m n_n n_o n_p - n_\alpha n_j n_k n_\ell\right)}$$

4 spin cross-relaxation

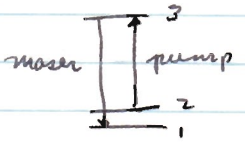
Cross-relaxation prevents maser action at very small frequencies, i.e, of the order of the linewidth.



Because of the cross-saturation, the populations of 2 and 3 will become equal. This is the reason why <u>diluted</u> paramagnetic solids only can have maser action.

Maser action has been observed in the following case: — 4

This only happens for $\nu_{41} = 2\nu_{32}$



$W_{pump}(\nu_{32})$ causes $n_3 = n_2$

$W_{cross}(n_3^2 n_1 - n_2^2 n_4)$ causes $n_1 = n_4$

Thus allowing maser action on ν_{31}.

3 MAY 1963

Transient Behaviour of Maser Oscillators

Consider a 2 level system whose energy lies near a cavity mode. We treat the field classically. Essentially, the dynamical variables of the problem are the diagonal and off-diagonal elements of the density matrix. Recall:

$$(1)\quad i\hbar\,\dot{\rho}_{ba} = \mathcal{H}^{int}_{ba}(\rho_{aa}-\rho_{bb}) + \hbar\omega_{ba}\rho_{ba} - i\hbar T_2^{-1}\rho_{ba}$$

Take: $\mathcal{H}^{int}_{ba} = -\underset{\sim}{\mu}\cdot\underset{\sim}{E}$; $\mu = \mu_{ba} = \mu_{ab}$

Now:

$$\underset{\sim}{P}(\underset{\sim}{r},t) = \left(\underset{\sim}{\mu}_{ab}\rho_{ba}(t) + \underset{\sim}{\mu}_{ba}\rho_{ab}(t)\right) N(\underset{\sim}{r})$$

and: $$\underset{\sim}{P}(t) = \int \underset{\sim}{P}(\underset{\sim}{r},t)\,dV$$

Also:

$$(2)\quad \dot{\rho}_{bb} - \dot{\rho}_{aa} = \frac{2}{i\hbar}\left(\rho_{ab}\mathcal{H}^{int}_{ba} - \rho_{ba}\mathcal{H}^{int}_{ab}\right) - \frac{\rho_{bb}-\rho_{aa}-\rho^{\circ}_{bb}+\rho^{\circ}_{aa}}{T_1}$$

For the field: $\underset{\sim}{E}(\underset{\sim}{r},t) = -\sqrt{4\pi}\sum_\lambda p_\lambda(t)\,\underset{\sim}{E}_\lambda(\underset{\sim}{r})$
and:

$$(3)\quad -\sqrt{4\pi}\left\{\ddot{p}(t) + \frac{\omega_\lambda}{Q}\dot{p}(t) + \omega_\lambda^2 p(t)\right\} = -\frac{4\pi}{\epsilon\mu}\,\frac{\int \frac{\partial^2 \underset{\sim}{P}(\underset{\sim}{r},t)}{\partial t^2}\cdot\underset{\sim}{E}_\lambda(\underset{\sim}{r})\,dV}{\int \underset{\sim}{E}_\lambda(\underset{\sim}{r})\cdot\underset{\sim}{E}_\lambda(\underset{\sim}{r})\,dV}$$

Note this is a non-linear equation because $\underset{\sim}{P}$ depends on ρ_{ab} and hence has no general closed solution.

The unperturbed material system energy is:

$$W = \frac{1}{2} \hbar \omega_{ba} (\rho_{bb} - \rho_{aa}) N$$

Multiply (1) by $\hbar \omega_{ba} \mu \int N(\underline{r}) dV$:

$$\imath \hbar^2 \omega_{ba} \dot{\rho}_{ba} \mu \int N(\underline{r}) dV = \mu^2 E \hbar \omega_{ba} (\rho_{aa} - \rho_{bb}) \cdot \int N(\underline{r}) dV + \hbar^2 \omega_{ba} \dot{\rho}_{ba} \mu \int N(\underline{r}) dV$$

Similarly for $(\ast\ast)^T$, multiply by $\hbar \omega_{ab} \mu \int N(\underline{r}) dV$; then add results. Note we have left out damping. We use:

$$\dot{\underline{P}}(\underline{r}, t) = -\imath \mu \omega_{ba} \rho_{ba} - \imath \mu \omega_{ab} \rho_{ab}$$

and the relation for W, resulting in:

$$(1)' \quad \ddot{P} + \omega_{ba}^2 P + \frac{\dot{P}}{T_2} = -4 \mu^2 \hbar^{-2} W E$$

Similar operations on (2) yields:

$$(2)' \quad \dot{W} = E \dot{P} - \frac{(W - W_0)}{T_1}$$

and (3) gives:

$$(3)' \quad \ddot{E} + \frac{\omega_\lambda}{Q} \dot{E} + \omega_\lambda^2 E = \frac{4\pi \omega^2 P}{\epsilon \mu} \eta$$

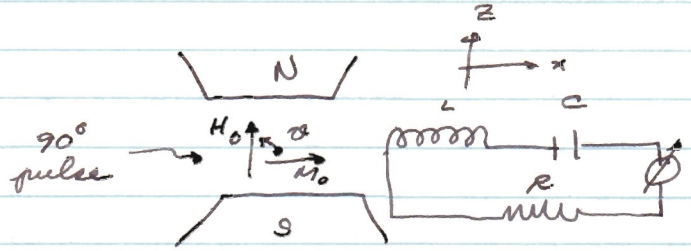
Take the case: $T_1 = T_2 = Q = \infty$. Energy should now be conserved. Operate on (3)' with $\int \dot{E} \, dt$:

$$\frac{1}{2} \dot{E}^2 + \frac{1}{2} \omega_\lambda^2 E^2 = \frac{4\pi \omega^2}{\epsilon \mu} \eta PE - \frac{4\pi \omega^2}{\epsilon \mu} \eta W + \text{constant}$$

or:

$$\underbrace{\dot{E}^2 + \omega_\lambda^2 E^2}_{\text{field}} + \underbrace{\frac{8\pi \omega^2 \eta}{\epsilon \mu} (W - P\dot{E})}_{\text{material}} = \text{constant}$$

Consider:



$M_z = M_0 \cos\theta$

$M_x = M_0 \sin\theta \sin\omega_0 t$

$M_y = M_0 \sin\theta \cos\omega_0 t$

Energy dissipated in resistance comes from initial orientation of M. Teng' saw reaction tends to return M to H_0. We now calculate how long it takes this reaction to return M to H_0. The voltage induced in C is:

$$V_{ind} = -n A \eta \frac{4\pi}{c} \frac{dM_x}{dt}$$

(turns, area)

$$= n A \eta \frac{4\pi\omega_0}{c} M_0 \sin\theta \cos\omega_0 t$$

$$I_{ind} = \frac{V_{ind}}{R} = \frac{VQ}{\omega L}$$

$$H_x = \frac{4\pi n}{cl} I_{ind}$$

Then:

$$\frac{dM_z}{dt} = -\gamma M_y H_x$$

$$= 2\pi\eta M_0^2 Q \gamma \sin^2\theta = -M_0 \sin\theta \frac{d\theta}{dt}$$

or:

$$\frac{d\theta}{dt} = -2\pi\eta M_0 Q \gamma \sin\theta$$

or:

$$\frac{1}{\cos^2\frac{\theta}{2}} \left(\frac{d\theta}{\tan\frac{\theta}{2}}\right) = -\frac{dt}{T_R}$$

or: $\tan\frac{\theta}{2} = \exp\left(-\frac{t}{T_R}\right)$; $T_R = (2\pi\eta M_0 Q \gamma)^{-1}$

For this to hold: $T_1 \gg T_R$, and must have heavy damping in circuit.

6 MAY 1963

Recapitulation:

$$i\hbar\,\dot\rho_{ba} = \mathcal{H}_{ba}(\rho_{aa}-\rho_{bb}) + \hbar\omega_{ba}\rho_{ba} - i\hbar T_2^{-1}\rho_{ba}$$

$$\dot\rho_{bb} - \dot\rho_{aa} = \frac{2}{i\hbar}(\rho_{ab}\mathcal{H}_{ba} - \mathcal{H}_{ab}\rho_{ba}) - \frac{\rho_{bb}-\rho_{aa}+\rho^0_{aa}-\rho^0_{bb}}{T_1}$$

$$-\sqrt{4\pi}\left(\ddot\rho_\lambda(t) + \frac{\omega_\lambda}{Q}\dot\rho_\lambda + \omega_\lambda^2\rho_\lambda\right) = -\frac{4\pi}{\epsilon}\frac{\int \frac{\partial^2 P}{\partial t^2}\cdot \underset{\sim}{E}_\lambda\, dV}{\int \underset{\sim}{E}_\lambda\cdot\underset{\sim}{E}_\lambda\, dV}$$

$$\mathcal{H}_{ba} = \mathcal{H}_{ab} = -\mu E$$

From these equations, we obtain:

$$\ddot P = \mu(\ddot\rho_{ab}+\dot\rho_{ba}) = \frac{1}{i}\mu\omega_{ba}(\dot\rho_{ba}-\dot\rho_{ab})N$$

$$\ddot P = \frac{2\mu^2 E}{\hbar}\omega_{ba}(\rho_{aa}-\rho_{bb})N - \omega_{ba}^2(\rho_{ba}+\rho_{ab})\mu N$$

Now: $W_0 = -\frac{1}{2}\hbar\omega_{ba}(\rho_{aa}-\rho_{bb})N$

and the first equation gives:

$$\ddot P + \omega_{ba}^2 P = -4\mu^2\hbar^{-2}E W_0 \qquad \left(\frac{\omega_{ba}}{Q=T_2}\dot P \text{ to LHS}\right)$$

the second gives: $\dot W = E\dot P$ $\qquad \left(-\frac{W-W_0}{T_1} \text{ to RHS}\right)$

and the third:

$$\ddot E + \omega_\lambda^2 E = \frac{4\pi\omega^2}{\epsilon}P\eta \qquad \left(\frac{\omega_\lambda}{Q}\dot E \text{ to LHS}\right)$$

Recall that our magnet problem of last time gave:

case (a): $\frac{\omega}{Q} \ll \gamma_R \ll T_2$; $T\gamma_R = (4\pi M_0 Q\eta)^{-1}$

For case (b), we consider only small changes in the population so that W is approximately constant.

Hence, the first two equation become linear. This corresponds to M originally parallel to H_0 with a very small current in the circuit. The second equation gives $\dot{W} = 0$ so that the first and third give:

$$\left(-\omega^2 + i\frac{\omega}{T_2} + \omega_{ba}^2\right) P + 4\pi\mu^2\hbar^{-2} W_0 E = 0$$

$$-\frac{4\pi\omega^2}{\epsilon}\eta P + \left(-\omega^2 + i\frac{\omega_d\omega}{Q} + \omega_d^2\right) E = 0$$

For the case of no damping, the characteristic equation gives:

$$\omega^2 = \frac{\omega_{ba}^2 + \omega_d^2}{2} \pm \sqrt{\frac{(\omega_{ba}^2 + \omega_d^2)^2}{4} - \omega_d^2\omega_{ba}^2 - \frac{16\pi\omega_d^2\eta\mu^2\hbar^{-2}W_0}{\epsilon}}$$

Note that an approximation has been made:

$$-\frac{4\pi\omega^2}{\epsilon}\eta P \rightarrow -\frac{4\pi\omega_{ba}^2}{\epsilon}\eta P$$

For: $\omega_d \sim \omega_{ba}$:

$$\omega = \omega_{ba}\left[1 \pm \underbrace{\frac{\sqrt{-\frac{16\pi}{\epsilon}\eta\mu^2\hbar^{-2}W_0}}{2\omega_{ba}}}_{\sim 10^{-6}}\right]$$

This justifies our initial of small coupling to the external circuit.

Now define:

$$\dot{E} - i\omega E = -i\omega E' e^{-i\omega_{ba}t}$$

$$\rho_{ba} = \rho'_{ba} e^{-i\omega_{ba}t}$$

It follows that:

$$\ddot{E} + \omega_{ba}^2 E = i\omega_{ba}\dot{E}' e^{-i\omega_{ba}t}$$

and:

$$\imath \omega_{ba} \dot{E}' + \underbrace{(\omega_c^2 - \omega_{ba}^2)}_{0 \text{ for } \omega_c = \omega_{ba}} E e^{\imath \omega_{ba} t}$$

$$= \frac{4\pi \omega^2 \mu}{\epsilon} \eta \left(\rho'_{ba} + \underbrace{\rho'_{ab} e^{2\imath \omega_{ba} t}}_{=0 \text{ because it is a non-secular term, very rapid oscillation with small amplitudes}} \right)$$

Case (C): T_2 very short compared to interaction with circuit, i.e., material strongly damped so that steady state is assumed almost immediately. This gives:

$$\imath \hbar \dot{\rho}'_{ba} = -\imath \hbar T_2^{-1} \rho'_{ba} - \mu E (\rho_{aa} - \rho_{bb})$$

and we must add to the LHS of the E' equation:

$$\frac{\imath \omega_{ba}^2}{2Q} E'$$

Then:

$$\imath \omega_{ba} \dot{E}' + \frac{\imath \omega_{ba}^2}{2Q} E' = \frac{4\pi \omega^2 \mu^2}{\epsilon} \eta E' (\rho_{bb} - \rho_{aa}) \cdot T_2 N \hbar^{-1}$$

or; forming:

$E' \dot{E}'^* - E' \dot{E}'^*$, etc, we find:

$$\underbrace{\frac{\epsilon}{4\pi} \frac{d}{dt} |E'|^2}_{\text{increase per unit time in stored energy}} + \underbrace{\frac{\epsilon}{4\pi} \frac{\omega_{ba}}{Q} |E'|^2}_{\text{energy dissipated in cavity losses}}$$

$$= \underbrace{2\hbar \omega_{ba} \mu^2 \eta |E'|^2 (\rho_{bb} - \rho_{aa}) T_2 N \frac{1}{\hbar^2}}_{\text{Negative power absorbed by atomic system}}$$

This case fits the solid state optical masers.

In terms of number of photons:

$(\rho_{bb} - \rho_{aa}) N = 2n$ is the surplus in the upper state.

$$\frac{d n_{ph}}{dt} = k \eta n_{ph} - \frac{\omega}{Q} n_{ph}$$

$$\frac{d n}{dt} = - k \eta n_{ph} - \frac{n - n_0}{T_1}$$

For no damping, note:

$$\frac{d}{dt} (n_{ph} + n) = 0$$

and is a conserved quantity

8 MAY 1963

We have considered the case of large material system damping, so that (short T_2) the material system comes to steady state immediately.
Let the population difference be n such that:

$$n = N(\rho_{bb} - \rho_{aa})$$

from which:

$$\frac{dn}{dt} = -2k\eta n_{ph} - \frac{n - n_0}{T_1}$$

$$\frac{dn_{ph}}{dt} = k\eta n_{ph} - \frac{\omega}{Q} n_{ph}$$

Let us assume for the steady state:

$$n = n_0 + \Delta n + n^{eq}$$
$$n_{ph} = n_0 + \Delta n$$

and neglect mixed terms in the Δ's.

$$\frac{d(\Delta n)}{dt} = -2k\eta_0 n_0 + R_{pump} - \frac{\Delta n}{T_1} - 2k n_0 \Delta\eta - 2k\eta_0 \Delta n - \frac{n^{eq} - n_0}{T_1}$$

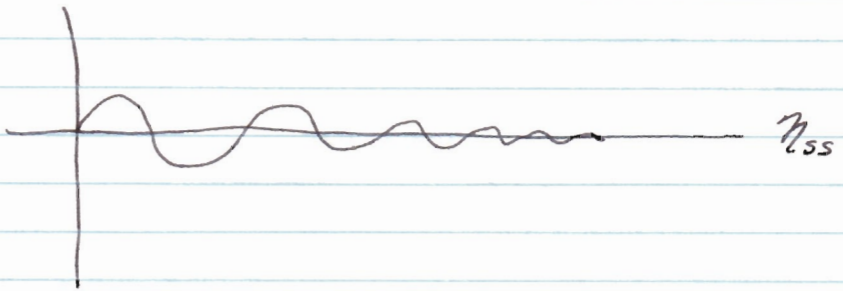
$$\frac{d(\Delta n)}{dt} = \left(k\eta_0 - \frac{\omega}{Q}\right) n_0 + \left(k\eta_0 - \frac{\omega}{Q}\right)\Delta n + k n_0 \Delta\eta$$

Note that the originally steady state terms are included and are zero, so that we are just considering small deviation from the steady state.

Assuming $e^{i\omega_{real} t}$ as a solution, we obtain the secular equations; and the secular determinant:

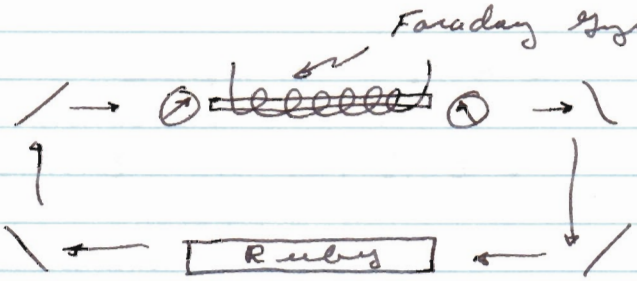
$$\begin{vmatrix} (\iota\omega_{rel} + \frac{1}{T_1} + 2kn_0) & 2k\eta_{ss} \\ -kn_0 & \iota\omega_{rel} + k\eta_{ss} - \frac{\omega}{Q} \end{vmatrix} = 0$$

which gives a behaviour:



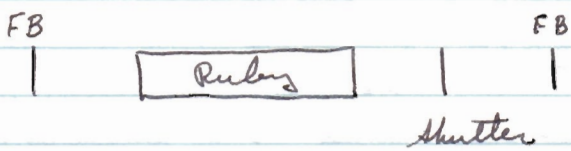
This behaviour is actually observed in microwave masers. For lasers, Δn is not necessarily small and we must consider more than one mode.

However, we may form a traveling wave laser:



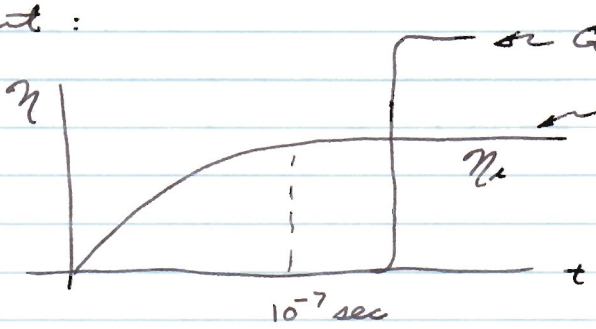
This configuration permits semidirectional flow of light. The traveling wave laser gives a very uniform spiking.

Consider now the Q-switched laser:



Without shutter, Ruby gain with 52% of Cr in upper state and 48% in ground state. With shutter, one can pump much higher without oscillation. If shutter is suddenly opened, oscillation occurs in 5 or 6 passes and one gets a "giant" pulse. The shutter is actually a Glan-Thompson polarizer and a Kerr cell. This can be switched in a microsecond.

We now consider the mathematical treatment:

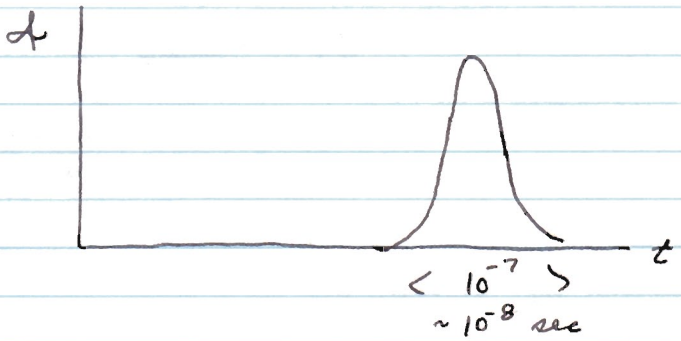


or Q when Kerr cell switched

or equilibrium due to weak laser action

note: $\frac{n - n_0}{T_1}$ is essentially 0.

The light output looks like:



We have considered the pump to be on continuously (actually ~ 10^{-3} sec). We have then:

$$\frac{d \ln n}{dt} = -2kn$$

$$\text{or:} \quad n = n_i \, e^{-2k \int_0^t n \, dt'}$$

Hence:

$$\frac{dn}{dt} = k\eta_a e^{-2k\int_0^t n\,dt'} - \frac{\omega}{Q} n$$

Rewrite as:

$$\frac{d^2v}{dt^2} = k\eta_a e^{-2kv}\frac{dv}{dt} - \frac{\omega}{Q}\frac{dv}{dt}$$

and:

$$\frac{dv}{dt} = \tfrac{1}{2}\eta_a\left(1-e^{-2kv}\right) - \frac{\omega}{Q}v + C''$$

and then:

$$t = \int_0^v \frac{dv}{\tfrac{1}{2}\eta_a\left(1-e^{-2kv}\right) - \frac{\omega}{Q}v + C''}$$

and the problem is solved in principle. We need to know $n(t)$. We know:

$$\left(\frac{dn}{dt}\right)_{t=0} = \frac{1}{2}\left(N^{\text{Total}} - \eta_a\right) W_{\text{pump}}\,\xi$$

(n initial) (ξ: fluorescence efficiency)

$$= \left(k\eta_a - \frac{\omega}{Q}\right) n_{t=0}$$

Note then: $\left(\frac{dv}{dt}\right)_{t=0} = C'' = n_{t=0}$

If we ignore $\frac{\omega}{Q}v$ (can be in initial stages of build-up) we can get:

$$t = \int_0^v \frac{d\left(e^{2kv}\right)}{\left(\frac{1}{2}\eta_a + C''\right)e^{2kv} + \frac{1}{2}\eta_a}$$

$$= \frac{1/2k}{\frac{1}{2}\eta_a + C''} \ln\left(\frac{e^{2kv} + \frac{\frac{1}{2}\eta_a}{\frac{1}{2}\eta_a + C''}}{1 + \frac{\frac{1}{2}\eta_a}{\frac{1}{2}\eta_a + C''}}\right)$$

now note:

$$e^{2k\nu} = \frac{n_2}{n(t)}$$

The characteristic time of build-up, τ_{bu} is:

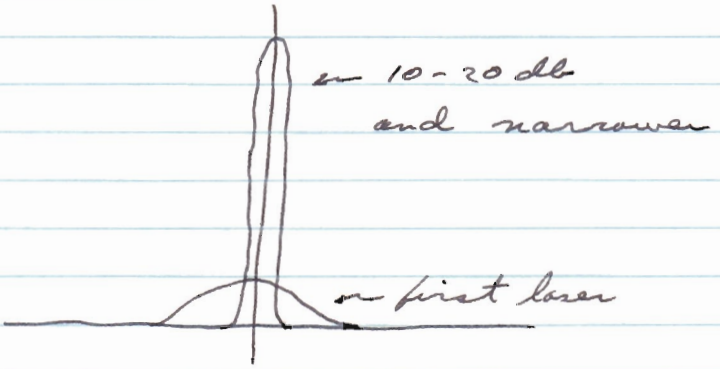
$$\tau_{bu} = \frac{t_{\text{single pass}}}{\alpha_2 \, l}$$

where α_2 is the initial gain per pass.

The result in that we have a device which delivers tremendous amounts of power for a very short time. Take 5cc of ruby, for which $n_2 \approx 2.5 \cdot 10^{19}$. Each quantum has $h\nu = (6.62 \cdot 10^{-27})(4 \cdot 10^{14}) = 2.5 \cdot 10^{-12}$ ergs. The total energy is $\sim 6 \cdot 10^7 \sim 6$ joules. Actually we get .1 to 1 joule out, but this is delivered in $\sim 10^{-8}$ sec. This is a power of 10 - 100 megawatts/cm^2 peak power. This can be amplified by feeding the output to:

→ [Ruby] ← antireflecting coatings

One gets a higher peak power and a narrower pulse.



This gives about 1000 megawatts peak power.

The analysis of the second ruby must include a diffusion term because of the change in the density of states due to the very intense light:

$$\frac{d\,n(x,t)}{dt} = k\,\eta_r(x)\,n\,e^{-2k\int_0^t n\,dt'} - c\,\frac{\partial n}{\partial x}$$

note this does not increase the total energy very much but is a power peaking device.

10 MAY 1963

Consider again the Q switched laser: Take:

$$E_{light} = 30 \text{ kv/cm} = 100 \text{ esu}$$

Then: $$\frac{cE^2}{4\pi} = \frac{3\cdot 10^{10}\cdot 10^4}{4\pi} = 2.5\cdot 10^6 \text{ watts/cm}^2$$

so this gives us an idea of the E field involved. One can increase the flux by focusing. This makes the field about that inside the atom and we should expect to observe non-linear effects.

Non-linear effects are not new, in fact, they are covered by Maxwell's equations. Note $D = \epsilon E$ and $B = \mu H$ in ferro-type materials.

We will outline how to calculate non-linear susceptibilities. Assume:

$$\underset{\sim}{D} = \underset{\sim}{E} + 4\pi \underset{\approx}{\chi}\,\underset{\sim}{E} + 4\pi \underset{\approx\approx}{\chi}^{NL}\,\underset{\sim}{E}\cdot\underset{\sim}{E} + \cdots$$

(Note: $E_{atomic} \sim 3\cdot 10^8$ kv/cm)

Usually, the nonlinearity will be small and we will be concerned with the complex $\underset{\approx\approx}{\chi}^{NL}$. They will be two approaches, the first being to calculate $\underset{\approx}{\chi}^{NL}$ quantum mechanically, the second then being to take this phenomenologically as a given quantity of a material, veg:

$$\underset{\sim}{D} = \underset{\approx}{\epsilon}\,\underset{\sim}{E} + 4\pi \underset{\sim}{P}^{NLS}$$

and then the treatment of EM propagation in such a medium.

Let us first consider the classical problem of the slightly anharmonic oscillator.

Then; consider:

$$m\ddot{x} + m\Gamma\dot{x} + m\omega_0^2 x + \alpha' x^2 = Re\left\{ eE\, e^{i\vec{k}\cdot\vec{r} - i\omega t} \right\}$$

Try a Fourier series solution: $x = \sum_m x_m e^{-im\omega t}$

We know the linear solution is:

$$P^1 = e\,x_{lin} = Re\ \frac{e^2 E\, e^{i\vec{k}\cdot\vec{r} - i\omega t}}{m(-\omega^2 + \omega_0^2 - i\omega\Gamma)}$$

For a non-dense medium: $P = N_0 P^1$

Proceeding:

$$x_2(2\omega) = \frac{\alpha'\left(\frac{e}{m}\right)^2 E^2\ e^{2i\vec{k}\cdot\vec{r} - 2i\omega t}}{(-\omega^2 + \omega_0^2 - i\omega\Gamma)^2(-4\omega^2 + \omega_0^2 - 2i\omega\Gamma)}$$

Then:

$$P^{NL}(2\omega) = Re\ \frac{\alpha'\left(\frac{e}{m}\right)^3 E^2\ e^{2i\vec{k}\cdot\vec{r} - 2i\omega t}}{(-\omega^2 + \omega_0^2 - i\omega\Gamma)^2(-4\omega^2 + \omega_0^2 - 2i\omega\Gamma)}$$

For a non-dense medium:

$$\chi = \frac{N_0}{4\pi}\ \frac{\alpha'\left(\frac{e}{m}\right)^3}{(-\omega^2 + \omega_0^2 - i\omega\Gamma)^2(-4\omega^2 + \omega_0^2 - 2i\omega\Gamma)}$$

$$+ \text{ DC term}$$

Also, if we apply 2 fields at ω_1 and ω_2, we will form the appropriate beat frequencies.

Our above treatment will be found to be strongly suggestive of the atomic problem.

Consider the case of non-linearity in the free electron in a magnetic and electric field:

$$E_x = E_0 \cos(kz - \omega t)$$
$$H_y = H_0 \cos(kz - \omega t)$$

$$m\ddot{x} = eE_x - \frac{e}{c} v_z H_y$$
$$m\ddot{y} = 0$$
$$m\ddot{z} = \frac{e}{c} v_x H_y$$

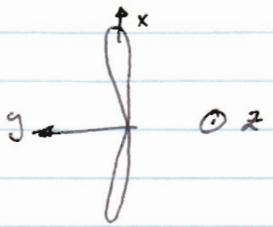
In first approximation, we say $\frac{v}{c} \ll 1$, and consider only the E field and get:

$$ex = -\frac{e^2}{m\omega^2} E_0 \cos\omega t$$

In the next approximation: (Use $\dot{x} = \frac{e}{m\omega} E \sin\omega t$)

$$z = -\frac{1}{8} \frac{eH_0}{mc\omega} \frac{eE_0}{m\omega^2} \sin(2kz - 2\omega t)$$

The motion in the z direction looks like:



The elongation is proportional to $\frac{\frac{eH_0}{mc}}{\omega} = \frac{\omega_c}{\omega}$

For the Q pumped laser, $H_0 \approx 100$ esu:

$\frac{\omega_c}{\omega} = \frac{2.8 \cdot 10^8}{4 \cdot 10^{14}} \approx 10^{-6}$ so that the non-linearity is very small.

Another non-linear case is the magnetic dipole in an EM field. Recall:

$$\dot{M}_x = \gamma M_y H_0 - \frac{M_x}{T_2}$$

$$\dot{M}_y = -\gamma M_x H_0 + \gamma M_z H_x \cos\omega t - \frac{M_y}{T_2}$$

$$\dot{M}_z = \gamma M_y H_x \cos\omega t - \frac{M_z - M_0}{T_1}$$

It is seen that these equations are non-linear and will lead to second harmonic terms.

References: IEEE, Jan '63, Bloembergen
RMP, Jan '63, Franchen and Ward.

13 MAY 1963

Recall:

$$m\ddot{x} + m\omega_0^2 x + m\Gamma\dot{x} + \alpha' x^2 = e E_{loc} \cos(\underline{k}\cdot\underline{r} - \omega t)$$

$$= \tfrac{1}{2} e E_{loc} \left[e^{i\underline{k}\cdot\underline{r} - i\omega t} + e^{-i\underline{k}\cdot\underline{r} - i\omega t} \right]$$

from which we deduce:

$$x_2(2\omega) = \frac{\tfrac{1}{4}\alpha' e^2 E_{loc}^2 \, e^{2i\underline{k}\cdot\underline{r} - 2i\omega t}}{\left(-m\omega^2 + m\omega_0^2 - i\omega m\Gamma\right)^2 \left(-4m\omega^2 + m\omega_0^2 - 2i\omega m\Gamma\right)}$$

$$+ CC$$

From this, one can define a complex, non-linear polarizibility, viz:

$$P^{NL}(2\omega) = \beta^{NL} \tilde{E}_{loc} \, e^{2i\underline{k}\cdot\underline{r} - 2i\omega t}$$

It is hard to interpret the real and imaginary parts because there is no applied field at twice the frequency. In fact, the real part of the complex polarizibility can mean absorption if we think of a field at 2ω but $90°$ out of phase. Note:

(1) If P^{NL} leads $E(2\omega)$ in phase, there is work done on the wave.

(2) If P^{NL} lags $E(2\omega)$ in phase, there is extra absorbtion in the medium.

(3) If in phase, energy is stored in medium.

Thus there is no a priori means of assigning physical meaning to P^{NL}.

Change in energy stored due to non-linearity is $\frac{1}{2} \underset{\sim}{E}^*(2\omega) \cdot \underset{\sim}{P}^{NLS}(2\omega)$

Power absorbed per cycle due to non-linearity is:

$$\frac{1}{2}(2\omega)\int_{\text{cycle}} \underset{\sim}{E}^{*}\cdot\frac{\partial \underset{\sim}{P}^{NLS}}{\partial t}\,dt$$

We now repeat our calculations for β^{NL} with quantum mechanics. First consider more closely the connection with Maxwell's Equations. Recall:

$$\underset{\sim}{D} = \underset{\sim}{E} + 4\pi N_0 \underset{\sim}{P}^{L} + 4\pi N_0 \underset{\sim}{P}^{NL}$$

For a dense medium, one must consider the Lorentz corrections. We will restrict ourselves to cubic symmetry. Strictly, odd-rank tensors vanish in cubic crystals, and we must consider those structures that lack inversion symmetry. We know then:

$$E_{loc} = E + \frac{4\pi}{3}P^{L} + \frac{4\pi}{3}P^{NL}$$

$$P^{NL}(\omega_3) = \beta^{NL}(\omega_3 = \omega_1 + \omega_2)\, E_1^{loc} E_2^{loc} \cdot e^{i(\underset{\sim}{k}_1+\underset{\sim}{k}_2)\cdot\underset{\sim}{r} - i\omega_3 t}$$

There is now no non-linear effect at ω_1 because there is no field at ω_3. Hence:

$$E_1^{loc}(\omega_1) = \frac{\epsilon(\omega_1)+2}{3}E(\omega_1)$$

Then:

$$P^{NL}(\omega_3) = \beta^{NL}(\omega_3)\,\frac{\epsilon(\omega_1)+2}{3}\,\frac{\epsilon(\omega_2)+2}{3}E_1E_2$$

Now:

$$P^L(\omega_3) = \alpha \left[E_3(\omega_3) + \frac{4\pi}{3} P^L(\omega_3) + \frac{4\pi}{3} P_{NL}(\omega_3) \right]$$

(α: linear polarizibility)

or:

$$P^L(\omega_3) = \frac{\alpha}{1 - \frac{4\pi}{3}\alpha} E_3(\omega_3) + \frac{\frac{4\pi}{3}\alpha}{1 - \frac{4\pi}{3}\alpha} P^{NL}(\omega_3)$$

$$= \frac{\epsilon(\omega_3) - 1}{4\pi} E_3 + \frac{\epsilon(\omega_3) - 1}{3} P^{NL}(\omega_3)$$

Then:

$$D(\omega_3) = E_3 + \left[\epsilon(\omega_3) - 1\right] E_3 + 4\pi \left[\frac{\epsilon(\omega_3) - 1}{3} + 1 \right] P^{NL}(\omega_3)$$

$$= \epsilon(\omega_3) E_3 + 4\pi P^{NLS}$$

where:

$$P^{NLS} = \underbrace{\beta^{NL} \frac{\left[\epsilon(\omega_1) + 2\right]\left[\epsilon(\omega_2) + 2\right]\left[\epsilon(\omega_3) + 2\right]}{27} N_0}_{\chi^{NL}} E_1 E_2$$

We now have made the connection with the macroscopic theory. Consider:

$$\nabla \times \underset{\sim}{H} = \frac{1}{c} \frac{\partial \underset{\sim}{D}}{\partial t} = \frac{1}{c} \frac{\partial}{\partial t} \left(\underset{\sim}{\epsilon} \underset{\sim}{E} \right) + \frac{4\pi}{c} \frac{\partial \underset{\sim}{P}^{NLS}}{\partial t}$$

which leads to:

$$\nabla \times \nabla \times \underset{\sim}{E} + \frac{1}{c^2} \frac{\partial^2 \left(\underset{\sim}{\epsilon} \underset{\sim}{E} \right)}{\partial t^2} = \underbrace{-\frac{4\pi}{c^2} \frac{\partial^2 \underset{\sim}{P}^{NLS}}{\partial t^2}}$$

Can consider as source at ω_3

Note that at each point in crystal, we have a source at ω_3. The phase at these points depends on the sum of the phases of E_1 and E_2. One will always find points where phase leads and lags will cancel. Therefore, we must use crystals of size $(\vec{k}_1 + \vec{k}_2 - \vec{k}_3)\,l \sim \pi$. l is usually $\sim 10^{-2}$ cm.

15 MAY 1963

We consider the QM treatment of the non-linear susceptibility of a two level system:

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho]$$

$$\mathcal{H} = \mathcal{H}_0 + \underbrace{\mathcal{H}'(t)}_{\text{sinusoidal}} + \mathcal{H}^{\text{random}}(t)$$

We attempt a solution in ascending powers of $\mathcal{H}'$. We know that $\mathcal{H}^{\text{random}}$ can be replaced by a phenomenological damping term.

$$i\hbar\dot{\rho}_0 = [\mathcal{H}_0, \rho_0] + \text{damping terms} \qquad (0\text{th})$$

$$i\hbar\dot{\rho}_1 = [\mathcal{H}', \rho_0] + [\mathcal{H}_0, \rho_1] + \text{D.T.} \qquad (1\text{st})$$

$$i\hbar\dot{\rho}_2 = [\mathcal{H}', \rho_1] + [\mathcal{H}_0, \rho_2] + \text{D.T.} \qquad (2\text{nd})$$

Consider a two level system:

b ——— ↑ z, H_0

a ———

$$\omega_{ba} = \gamma\hbar H_0$$

$$\mu_{bb} = -\mu_0 \; ; \; \mu_{aa} = \mu_0 \; ; \; \mathcal{H}_0 = -\mu_z H_0$$

Take:

$$\mathcal{H}' = \mu_{ab}\left(H_1 e^{i\omega_1 t} + H_2 e^{i\omega_2 t}\right) + \mu_{ba}\left(H_1^* e^{-i\omega_1 t} + H_2^* e^{-i\omega_2 t}\right) + \left(H_3 e^{i\omega_3 t} + H_3^* e^{-i\omega_3 t}\right)(\mu_{zaa} + \mu_{zbb})$$

At first we take the diagonal element = 0. Then:

$$\dot{\rho}_{bb} - \dot{\rho}_{aa} = -2\,\frac{\mathcal{H}'_{as}\rho_{sa} - \rho^0_{as}\mathcal{H}'_{sa}}{i\hbar} - \frac{\rho_{bb} - \rho_{aa} - \rho^0_{bb} + \rho^0_{aa}}{T_1}$$

$$\dot{\rho}_{ab} = -i\hbar^{-1} \mathcal{H}'_{ab}(\rho_{aa} - \rho_{bb}) - \frac{\rho_{ab}}{T_2} + i\omega_{ba}\rho_{ab}$$

In zero approximation $\rho_{bb} - \rho_{aa} = \rho^{\circ}_{bb} - \rho^{\circ}_{aa}$

In first order:

$$\rho_{ab} = \frac{-i\hbar^{-1} M_{ab} H_1 (\rho^{\circ}_{bb} - \rho^{\circ}_{aa})}{i(\omega_1 - \omega_{ba}) + T_2^{-1}} e^{i\omega_1 t}$$

$$+ \text{ similar terms in } \omega_2$$

In second order, looking for DC terms only:

$$\rho^{dc}_{bb} - \rho^{dc}_{aa} = (\rho^{\circ}_{bb} - \rho^{\circ}_{aa}) \Bigg[1$$

$$- 2|M_{ab}|^2 |H_1|^2 \hbar^{-2} T_1 \frac{2T_2}{1 + (\omega_1 - \omega_{ba})^2 T_2^2} - 2|M_{ab}|^2 |H_2|^2 \hbar^{-2}$$

$$\cdot T_1 \frac{2T_2}{1 + (\omega_2 - \omega_{ba})^2 T_2^2} \Bigg] \quad \text{(incipient saturation terms)}$$

In addition:

$$(i\omega_3 + T_1^{-1})\left(\rho_{bb}^{(\omega_1 - \omega_2)} - \rho_{aa}^{(\omega_1 - \omega_2)}\right)$$

$$= 4\hbar^{-2} |M_{ab}|^2 H_1 H_2^* e^{i(\omega_1 - \omega_2)t} (\rho^{\circ}_{bb} - \rho^{\circ}_{aa})$$

$$\left\{ \frac{T_2}{i(\omega_1 - \omega_{ba})T_2 + 1} + \frac{T_2}{-i(\omega_2 - \omega_{ba})T_2 + 1} \right\} + \cdots$$

Then:

$$T_2 \langle M_z \rho \rangle^{(\omega_1 - \omega_2)} = M_0 (\rho_{bb} - \rho_{aa})^{(\omega_1 - \omega_2)}$$

$$= \frac{4\hbar^{-2} |M_{ab}|^2 H_1 H_2^* M_0 T_1 T_2}{(1 + i\omega_3 T_1)\{i(\omega_1 - \omega_{ba})T_2 + 1\}} e^{i(\omega_1 - \omega_2)t}$$

$$+ \cdots$$

$$+ cc + cc$$