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QUANTUM OPTICS: A HEURISTIC APPROACH - TERMINOLOGY AND NUMEROLOGY

(Taken from International School of Quantum Electronics)

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1 - Quantum Optics : a heuristic approach - Terminology and Numerology

1.1 Definition of Quantum Optics

The term "quantum electronics" was first used in 1959 at a conference dealing with the physical and engineering uses of the MASER (Microwave Amplifier by Stimulated Emission of Radiation) and MASER oscillators in high resolution spectroscopy and in the handling of electromagnetic signals at $\lambda \sim 1$ cm.

Half a year later, the first LASER was operated (L stays for light. The L has replaced the M because this time the generated radiation is at $\lambda \lesssim 1$ μm)

Since 1960 the LASER has become a useful device in many areas of physics and technology.

In the spectral range of interest, it is more convenient to speak of Quantum Optics, rather than Quantum Electronics.

Quantum Optics can be approached from three points of view, namely :

i) physics of the stimulated emission processes;

ii) coherence and cooperative phenomena in radiation -matter interaction;

iii) applications of the LASER related to its spectral purity.

We shall discuss the three aspects in sequence, defining the terms and giving the orders of magnitude.

1.2 Physics of the stimulated emission processes

If the e.m. cavity where we are considering the radiation–atom interaction is a rectangular cavity of sides X, Y, Z ; volume V = X Y Z , then the solution of the wave equation, with periodic boundary conditions, yields the plane wave expansion for the field

$$E(x, y, z, t) = \sum E_{\vec{k}}(t)\, e^{i(k_1 x + k_2 y + k_3 z)} \qquad (1.1)$$

where

$$k_1 = n_1 \cdot 2\pi / X$$

$$k_2 = n_2 \cdot 2\pi / Y \qquad (n_i = 1, 2, \ldots)$$

$$k_3 = n_3 \cdot 2\pi / Z$$

For each set of $k_{1,2,3}$ we have a different field configuration, or mode.

The dispersion relation imposes a constraint between frequency ω and amplitude $k = \sqrt{k_1^2 + k_2^2 + k_3^2}$ of the $\vec{k}$ vector

$$\omega = c\, k \qquad (1.2)$$

In $\vec{k}$ space (fig. 1.1) each mode occupies an elementary volume

$$\delta^3 k = \delta k_1\, \delta k_2\, \delta k_3 = \frac{(2\pi)^3}{V} .$$

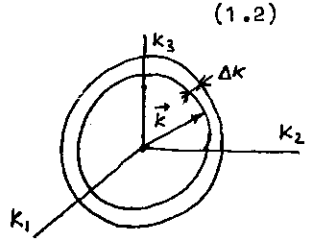


Fig. 1.1

In a spherical shell of radius k and thickness Δk there are

$$\Delta M = 2 \frac{4\pi k^2 \Delta k}{\delta^3 k} = \frac{k^2 \Delta k}{\pi^2} V \qquad (1.4)$$

modes. The extra-factor 2 accounts for the two possible polarizations for each $\vec{k}$ vector. Hence the mode density in frequency is $dM/d\omega = \omega^2 V/\pi^2$, or

$$\frac{dM}{d\nu} = \frac{8\pi\nu^2}{c^3} V \qquad (1.5)$$

If the cavity contains radiators (atoms on the walls or inside) in thermal equilibrium at a temperature T, then the electromagnetic energy density in the cavity is given by Planck's blackbody formula (1900)

$$\frac{dW}{d\nu} = \frac{8\pi\nu^2}{c^3} V \cdot h\nu \cdot \frac{1}{e^{h\nu/k_B T} - 1} = \frac{dM}{d\nu} \cdot h\nu \cdot \bar{n}_1(\nu) \qquad (1.6)$$

Here

$$\bar{n}_1(\nu) = \left(e^{h\nu/k_B T} - 1\right)^{-1}$$

is the average photon number for each mode whose $\vec{k}$ vector lies on the spherical surface of radius $k = 2\pi\nu/c$.

The distinction between spontaneous and stimulated emission came in the 1917 Einstein's derivation of Eq (1.6), as follows.

Consider two relevant levels of an atom coupled by an optical transition. Each time the atom goes up (absorption) or down (emission), this is a one-photon exchange process.

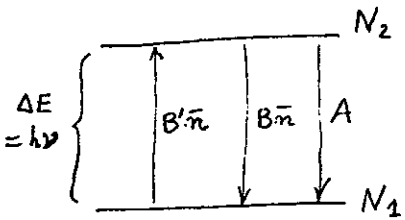


Fig. 1.2

The emission or decay process can be spontaneous (i.e., not triggered by photons) as well as stimulated (i.e., proportional to the photon number $\bar{n}$ at the frequency $\nu = \Delta E/h$); see fig. 1.2

If there is an ensemble of N atoms in thermal equilibrium, with N_2 in the upper state and $N_1 = N - N_2$ in the lower, then we have

$$N_1/N_2 = e^{\Delta E/k_B T} \qquad (1.7)$$

$$N_1 B' \bar{n} = N_2 B \bar{n} + N_2 A \qquad (1.8)$$

From this latter equation

$$\bar{n} = \frac{A/B}{\frac{B'}{B}\frac{N_1}{N_2} - 1}$$

By use of Eq (1.7) and comparison with (1.6), we get

$$\frac{A}{B} = \frac{8\pi\nu^3}{c^3} \quad , \tag{1.9}$$

$$B' = B \quad . \tag{1.10}$$

This can be interpreted by representing the degrees of freedom of the e.m. field as boxes and the excited atom as linked to all of them as in fig. 1.3

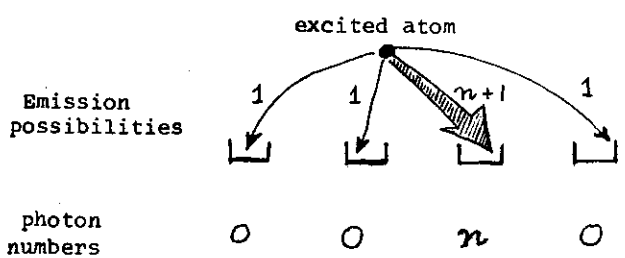


Fig. 1.3

With the probabilities there indicated, the stimulated emission probability into one mode is larger than the total spontaneous emission over ΔM modes (all those within the linewidth $\Delta\nu$ of the atomic emission) when

$$n > \Delta M. \tag{1.11}$$

In other words, it is not enough to have a mode with a large population ($n \gg 1$) but it is necessary to obey the specific condition (1.11) in order to observe stimulated effects. Hitherto we referred to the whole photon number in the cavity. It is more convenient to refer to the photon flux, given by $\Phi_n = c\, n / V$ or in terms of the field amplitude and the frequency ω, by

$$\Phi_n = \frac{c\,\varepsilon_0 E^2}{\hbar\omega} \qquad \left(\frac{\text{photons}}{\text{cm}^2\ \text{sec}}\right) \tag{1.12}$$

This becomes smaller for high frequencies ω, unless we simultaneously increase the E field strength.

The stimulated transition probability per second per atom is given by

$$p_s = \sigma \Phi_n \tag{1.13}$$

where the cross section σ for the process can be evaluated on a purely classical basis. For a free electron, it has the Thomson value

$$\sigma_{Th} = \frac{8}{3}\pi r_0^2 \sim 0.6 \times 10^{-24}\ \text{cm}^2 \tag{1.14}$$

where $r_0 = e^2/(4\pi\varepsilon_0 m c^2)$

is the classical electron radius.

For a bound electron, if the field frequency is resonant with the atomic transition, the cross section is

$$\sigma_{res} = \sigma_{Th}\left(\frac{\omega_0}{\gamma}\right)^2 \qquad (1.15)$$

At optical frequencies the decay rate γ can be as small as $10^{-5}\omega$, hence σ_{res} can be as large as $10^{10}\,\sigma_{Th}$, (fig. 1.4)

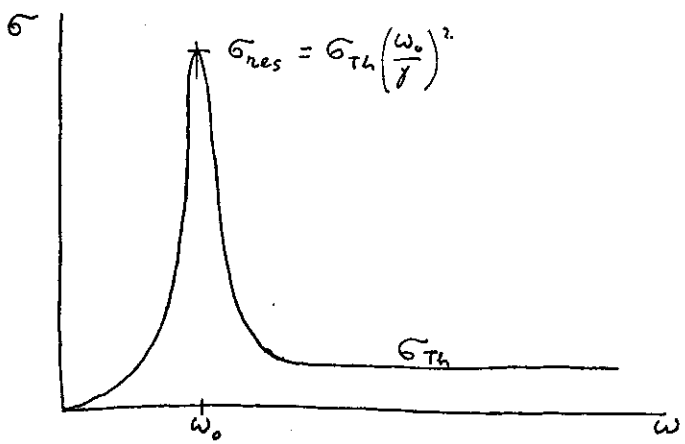


Fig. 1.4

Condition (1.11) with ΔM given by (1.4) is unfair. We try either to privilege some modes at constant σ, by a tuned cavity or to privilege a frequency by using the strong frequency dependence of σ_{res}.
The former is done at low frequencies (microwaves)

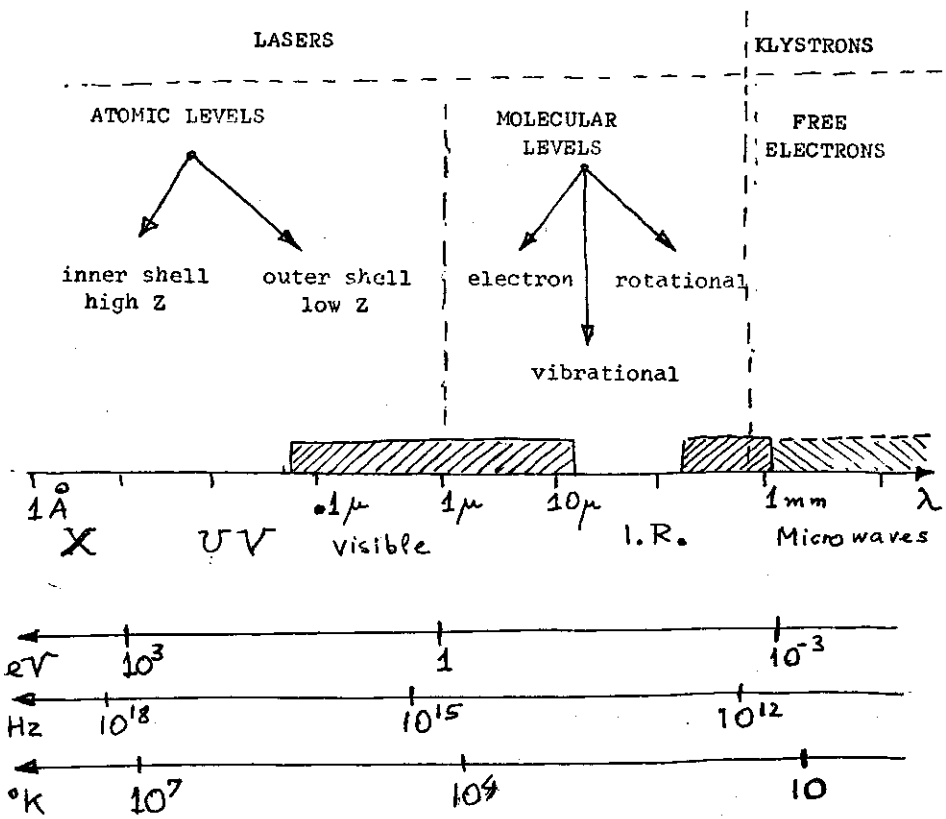


Fig. 1.5

the latter at high frequencies (from I R up)

By Eqs. (1.12) and (1.13), the condition (1.11) can be rephrased in terms of σ as $\sigma > \Delta M/\phi \propto \omega^3/E^2$. This shows that for high frequencies and a constant σ, stimulated emission can be obtained only by very high fields. Hence above 10^{12} Hz it is no longer convenient to extract energy from free electrons (with $\sigma = \sigma_{Th}$), even though outstanding examples are offered of e.m. fields in the visible and U.V. generated by free electrons (Cherenkov radiation, synchrotron radiation). In fig. 1.5 we report the regions where most lasers are available. It is useful to compare the λ scale with the eV, Hz and °K scales.

To put Eq (1.15) in a different way, we recall that if γ is due only to radiation damping (no collision broadening or other things), then from classical electro-dynamics

$$\frac{\omega}{\gamma} \sim \frac{\lambda}{\lambda_0}$$

hence

$$\sigma_{res} \sim \lambda^2/4\pi \qquad (1.15)'$$

Notice that the cross section is not the square of the electron radius nor the square of the Bohr's orbit but the squared wavelength which in the visible and UV is much bigger.

One arrives at the same numerical value by a semiclassical approach. Take the interaction Hamiltonian

$$\mathcal{H}' = -\vec{d}\cdot\vec{E} \qquad (1.16)$$

with a classical E field and a dipole operator whose matrix element between the two atomic states is $\langle 1|\, d \,|2\rangle = \mu$ ($\mu \sim 10^{-27}$ C x cm for an allowed transition). Then by Fermi's golden rule the transition rate is

$$W = \frac{2\pi}{\hbar^2}\left(\frac{dn}{d\omega}\right)_{max} \mu^2 E^2 \qquad (1.17)$$

putting $(dn/d\omega)_{max} = 2/(\pi\Delta\omega)$ (Lorentzian curve) and dividing by the photon flux $\phi_n = c\,\varepsilon_0 E^2/(\hbar\omega)$, one has

$$\sigma = \frac{W}{\phi_n} = \frac{4\omega}{\hbar c \varepsilon_0 \Delta\omega}\mu^2 \qquad (cm^2) \qquad (1.18)$$

This value is numerically as $\lambda^2/4\pi$.

Dividing w by the photon number in the cavity

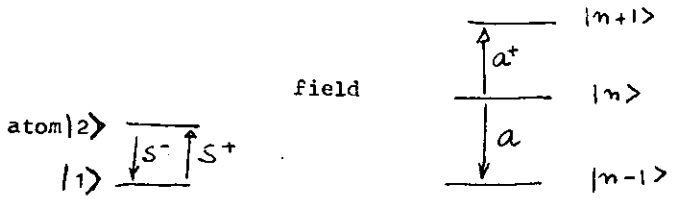
$$n = \varepsilon_0 E^2 V/(\hbar\omega) ,$$

one has the coupling constant per atom and per photon

$$g = \frac{1}{\hbar\varepsilon_0 V}\frac{\omega}{\Delta\omega}\mu^2 \qquad (1.19)$$

that we shall later use in the fully quantized interaction Hamiltonian (fig. 1.6)

$$H^1 = \hbar g\,(a^+ S^- + a\, S^+) \qquad (1.20)$$



$S^{\pm}$ = Pauli operators a^+ , a = Bose operators

Fig. 1.6

Let us now go back to condition (1.11). In order to keep a large photon number within the cavity, we must have small losses. This can be obtained by confining the radiating atoms within two mirrors which act as a photon store.

This way, the "ingredients" necessary for a laser are

i) an <u>active medium</u> i.e. a collection of atoms having a transition at the selected frequency

ii) a <u>pump</u> or excitation mechanism to take the atoms away from thermal equilibrium, up to excited state

iii) a cavity made of two facing mirrors with high reflectivity (this is called in optics a Fabry-Perot interferometer).

Such a cavity is resonant for those frequencies corresponding to the standing wave condition (fig .1.7)

$$m\frac{\lambda}{2} = L$$

which amounts to a minimum frequency separation

$$\Delta\nu_{m,m+1} = \frac{c}{2L} \qquad (1.21)$$

For these resonances, the escape time of photons is

$$T_d = \frac{L}{c}\,\frac{1}{1-R} \qquad (1.22)$$

that is, a transit time L/c multiplied by a loss factor $1/(1-R)$, R being the mirror reflectivity

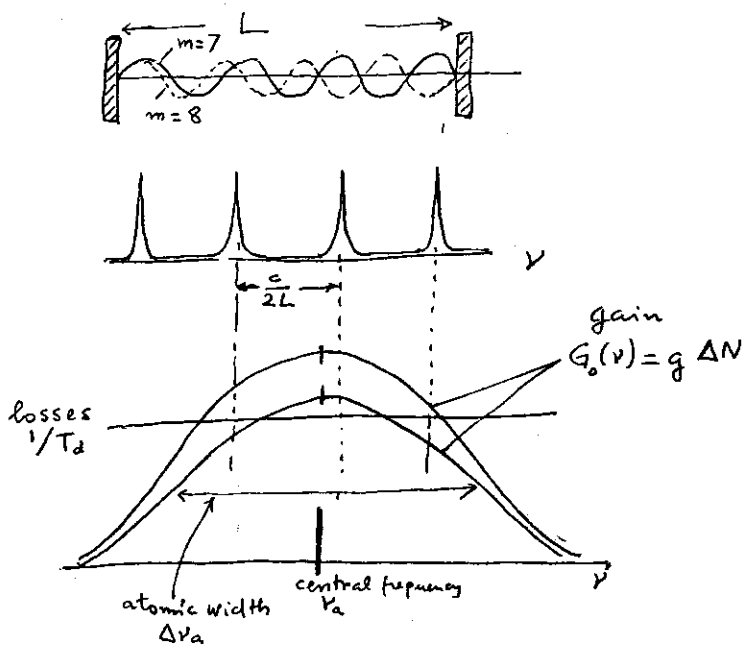


Fig. 1.7

Let us call $N = N_2 - N_1$, the difference of population between upper and lower state in the active medium, as <u>population inversion</u>.

Then, neglecting the spontaneous contributions (i.e. assuming Eq.(1.11) fulfilled) the rate equation for photons in the cavity will be

$$\frac{dn}{dt} = \frac{dn}{dt}\Big)_{gain} - \frac{dn}{dt}\Big)_{loss}$$

Or , by use of relations (1.19) and (1.22),

$$\frac{dn}{dt} = g \Delta N \cdot n - \frac{n}{T_d}$$

Hence the <u>threshold</u> condition for starting <u>oscillations</u> from an initial spontaneous <u>seed</u> (in other words: to have a self - sustained oscillator rather than an amplifier; so that we should say LOSER rather than LASER !) will be

$$g \Delta N \geq 1/T_d \qquad (1.23)$$

This condition is represented in Fig. 1.7 for two different pump values, i.e. for two different ΔN . In the first case only one mode is above threshold, hence we have a single monochromatic frequency. In the second case we may have emission at three frequencies. Here we must introduce the fundamental difference between <u>homogeneous</u> and <u>inhomogeneous</u> linewidth.

In the former case a monochromatic transition is broadened by circumstances which are <u>equal</u> for all atoms in the cavity (as spontaneous lifetime broadening, atomic collision broadening in a gas, phonon interaction in a solid matrix). All atoms can contribute over the <u>whole</u> linewidth. Hence, once the mode nearest to the peak has been excited, as the associated field "sweeps" the cavity, it will "eat" all atomic contributions, forbidding the other modes from going above threshold. In the standing wave case this frequency picture is not sufficient and one should also consider the space pattern. As sketched in fig. 1.8 two different modes have modes and maxima in different positions, hence they will "exploit" different atoms, releasing the competition. It is then possible the simultaneous laser action over many modes.

The inhomogeneous line broadening corresponds to different frequency locations of different atoms. This can be due, e.g., to Doppler shift in a gas where thermal agitation gives a distribution of velocities to the molecules and hence a distribution of Doppler shifts

$$\Delta\omega = \omega v_k / c$$

where V_k is velocity component along the $\vec{k}$ vector.

Another inhomogeneity occurs in a crystal where active ions are exposed to a crystal field which changes from site to site, contributing with a spread in the stark shifts.

For an inhomogeneous line, different modes can go above threshold even without a standing wave pattern.

In general if c/2L is much smaller than the atomic linewidth $\Delta\nu_a$ there are many independent laser lines, without phase relations.

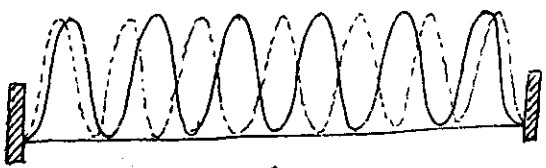


fig. 1.8

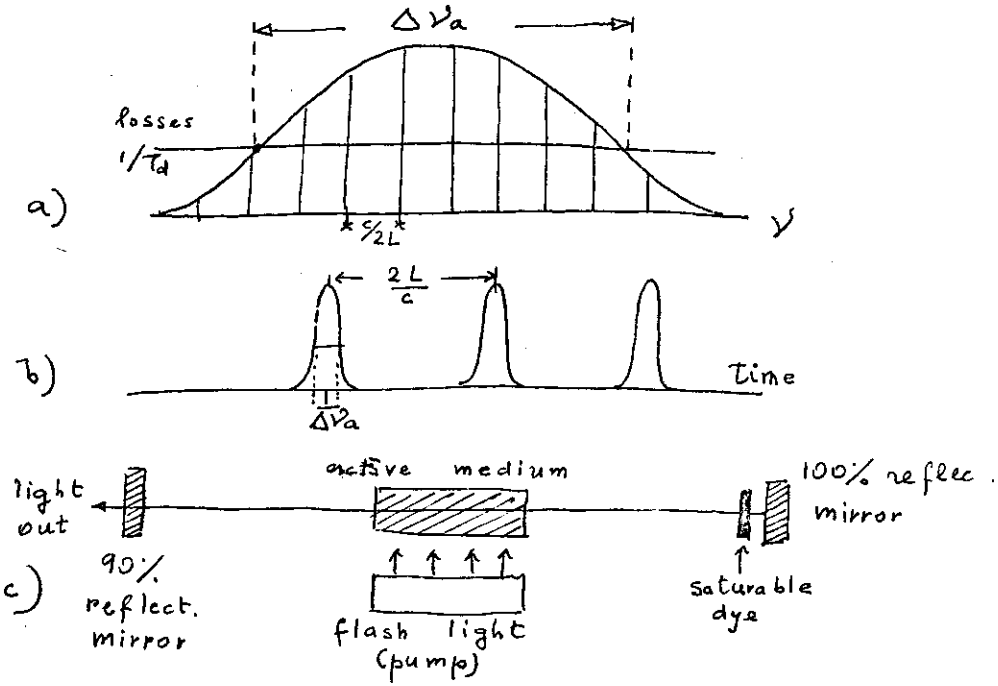


fig. 1.9 a - c

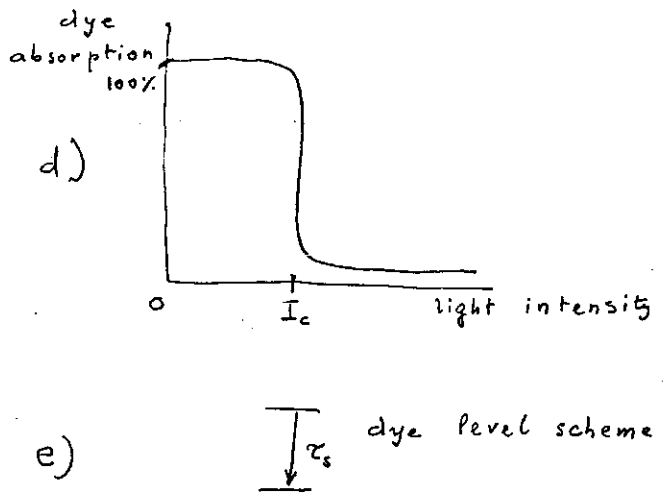


Fig. 1.9 Mode locking operation

a) frequency picture of a many-mode Laser

b) if the different laser fields have fixed phase relations, they act as the different Fourier components of a train of pulses, each lasting $1/\Delta\nu_a$ and separated by 2 L/c . b) is the Fourier transfer of the amplitude spectrum a), provided the phases are all equal

c) practical scheme of a mode-locked laser. Besides the three main ingredients (active medium, incoherent light to excite the atoms

at the upper level, mirrors) there is also a saturable dye which becomes transparent at a critical light intensity I_c (see d)). All the standing waves of the different modes will self-adjust their phases to have a maximum when the dye is transparent. Transparency is then lost with a decay time $\tau_s << 2L/c$ and then recovered after a transient 2 L/c. This corresponds to having a narrow pulse bouncing forth and back between the two mirrors. Notice that, from b) the pulse duration is $1/\Delta\nu_a$

In fig. 1.9 and in the associated caption it is explained how to lock in phase the laser lines, in order to make short pulses (as short as the uncertainty relation permits, i.e. $1/\Delta\nu_a$).

By using a Doppler broadened atomic line in a gas (like in a He-Ne, or in an A^+ laser), then

$$\Delta\nu \sim \frac{\nu}{c}\sqrt{\frac{k_B T}{M}} \sim 10^9 \text{ Hz}$$

hence

$$t_{pulse} \sim 1 \text{ n s} .$$

Using ions of a transition element imbedded in a crystal or glass matrix, as Cr^{3+} in Al_2O_3 (ruby), or Nd^{3+} in glass, one may have large $\Delta\nu_a$ because the 3 d or 4 f electron

is strongly affected by the crystal field. A large $\Delta\nu_a$ can also be achieved in the case of complex dye molecules in a liquid solution because of the overlapping among many vibrational and rotational levels. It is nowadays easy to achieve

$$\Delta\nu_a \sim 10^{13} \text{ Hz}$$

and hence

$$t_{pulse} \sim 0.1 \text{ p sec}$$

Notice that the range of picosecond times can be attained only by techniques as in fig. 1.7 , and not by electronic shutters.

1.3 Stimulated emission and nonlinear optics (NLO)

In fig. 1.3 we have represented the difference between spontaneous and stimulated emission. With reference to the Hamiltonian (1.20), and looking only at the field creation operator, it is well known that it acts on an m - photon state as follows

$$a^+ |n\rangle = \sqrt{n+1}\, |n+1\rangle . \qquad (1.21)$$

Therefore the transition rate for an emission process will be proportional to :

$$\text{spontaneous} : |\langle 1 | a^+ | 0\rangle|^2 = 1$$

$$\text{stimulated} : |\langle n+1 | a^+ | n\rangle|^2 = n+1$$

Also in higher order processes as those studied in NLO we can have a spontaneous and a stimulated version. Take a parametric process implying three Bose fields as in fig. 1. 10

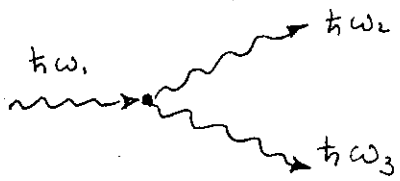


Fig. 1.10

The model Hamiltonian for the process in fig. 1.10 is

$$H^{I} = \hbar g \, (a_1 \, a^{+}_2 \, a^{+}_3 + a^{+}_1 \, a_2 \, a_3)$$

and the transition rate will have matrix elements as

$|\langle 1 | a^{+}_2 | 0 \rangle|^2 = 1$ spontaneous emission in the field 2

or

$|\langle n_2 + 1 | a^{+}_2 | n_2 \rangle|^2 = n_2 + 1$ stimulated emission in the field 2

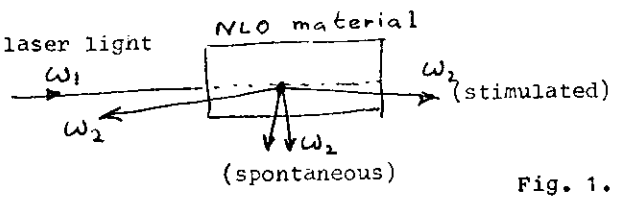


Fig. 1. 11

In the first case, we look at 90° , and we collect point-like processes, having to satisfy the conservation of energy:

$$\omega_1 = \omega_2 + \omega_3$$

In the second case, we look in a direction (forward or backward) almost collinear with the impinging beam. Here, in order to add coherently the field contributions, the momentum matching condition

$$\vec{k}_1 = \vec{k}_2 + \vec{k}_3$$

has also to be satisfied.

In a similar way we may describe usual light propagation in a transparent medium as an elastic two-photon process. Since the scattered contributions sum in phase, it is more convenient to speak of a linear polarization and a refraction index

$$P_i = \varepsilon_0 \chi^{(2)}_{ij} E_j \quad , \quad n_{ij} = \sqrt{\chi^{(2)}_{ij}} \qquad (1.22)$$

rather than stimulated emission in the scattered channel. Similarly, there are 4 photon processes leading to "self-actions" in the propagation of a large e.m. field, that is, self-focusing, self-defocusing, self modulation in phase (self-broadening) and amplitude (self-steepening). These non linearities on the same light beam are described by a nonlinear polarization index as

Table 1.1 - Examples of NLO processes

Nature of the quanta		Name of the process
2	3	
light	molecular vibrations	Raman
"	optical phonons in solids	"
"	acoustical phonons in solids	Brillouin
"	sound waves in liquids	"
"	light	parametric conversion (sum or difference of frequency, second harmonic generation etc.)

$$P_i = \chi^{(4)}_{ijkl} E_j E_k E_l \qquad (1.23)$$

The nonlinear refraction index can be written in the isotropic case as

$$n = n_0 + n_2 |E|^2 \qquad (1.23)^1$$

In a liquid of anisotropic molecules, self actions stem from orientation of the molecules due to interaction with the induced dipole moments (high frequency Kerr effect). In a liquid of isotropic molecules, or in solids and gases, self actions are due to electrostriction. For picosecond pulses, electrostriction is too slow to follow the amplitude change, and the effects are due to distortion of the electron cloud.

1.4 Coherence and cooperative phenomena

As shown in fig. 1.3, stimulated emission explains the privileged filling of a given mode, that is, a narrowing in the frequency spectrum and in the spectrum of possible directions (monochromaticity and directionality). This amounts to increasing the spectral purity , and use can be made of it in physics and technology (linear spectroscopy, holography, plasma production and compression by powerful laser pulses). But all this has very little to do with coherence .

Stimulated emission is a first order effect, i.e. it prov-

ides a linear source for Maxwell eqs.

Each mode has still a harmonic oscillator dynamics, that is, it is like a particle in a parabolic potential well, with an equilibrium statistical distribution given by Maxwell - Boltzmann. To have a sizeable amount of energy $|E_o|^2$ one has to increase the "temperature" i.e. the excitation, thus broadening the distribution and increasing the entropy as well (fig. 1.12).

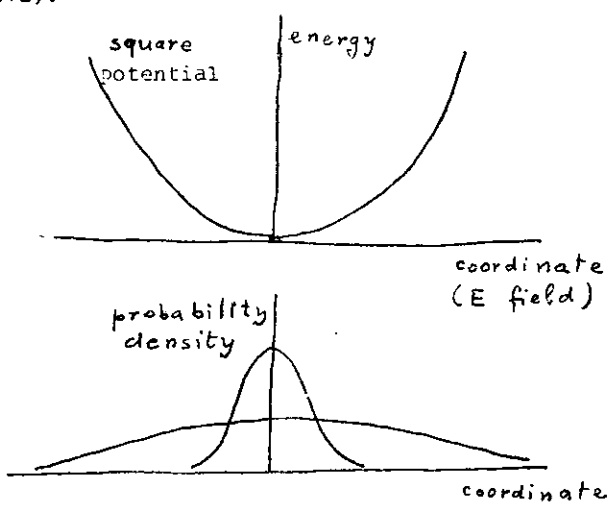


Fig. 1.12

However as the field E increases, one must consider high order processes, besides the one photon emission, as e.g. the three-photon process of fig. 1.13 which gives a quadratic field correction in the gain

$$G = G_o - \beta |E|^2 \qquad (1.24)$$

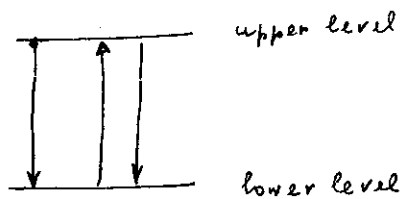


Fig. 1. 13

This is equivalent to a cubic polarization

$$P = G_o E - \tfrac{1}{3} \beta |E|^2 E$$

and to a quartic free energy

$$W = -P.E = -\frac{G_o}{2}|E|^2 + \frac{\beta}{42}|E|^4$$

As E increases, the quartic potential well becomes steeper and steeper (fig. 1.14), so that a useful E_o can be reached with a little amount of spread, or statistical fluctuations, around it.

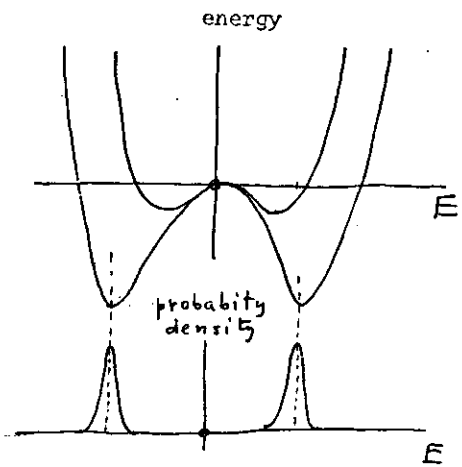


Fig. 1.14

We call coherent this highly excited field state without noise. The field can be described with very good approximation by a c-number with constant amplitude and phase. Such a field can bring the induced atomic dipoles to a coherent motion in which the phase relations among atomic wave functions are kept for long times.

This is the basis for a coherent nonlinear spectroscopy which sheds information on fine properties of atoms and molecules.