LASERS SOURCES *

F. T. Arecchi

University and
Istituto Nazionale di Ottica, Firenze

Since 1960 the Laser has become a useful device in many areas of physics and technology. A description of the laser can be done from two points of view, namely:

i) physics of the stimulated emission processes;
ii) coherence and cooperative phenomena in radiation-matter interaction.

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

1. 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_1, X_2, X_3, volume $V = X_1 X_2 X_3$, then the solution of the wave equation, with periodic boundary conditions, yields the plane wave expansion for the field

$$E(x, y, z, t) = \sum_{\vec{k}} E(\vec{k}, t)\, e^{i(k_1 x + k_2 y + k_3 z)} \qquad (1)$$

where $k_i = n_i \cdot 2\pi / X_i$ $(i = 1,2,3 \; ; \; n_i = 1,2,\ldots)$.

For each set of k_i we have a different field configuration, or mode.

The dispersion relation imposes a constraint between frequency ω and amplitude $k = (k_1^2 + k_2^2 + k_3^2)^{1/2}$ of the k vector

$$\omega = c\, k \qquad (2)$$

In k space each mode occupies an elementary volume

$$\delta^3 k = (2\pi)^3 / V. \qquad (3)$$

In a spherical shell of radius k and thickness Δk there

* Workshop on "IMAGING PROCESSES AND COHERENCE IN PHYSICS"
Les Houches - 12-23 March 1979

are

$$\Delta M = 2 \frac{4\pi k^2 \Delta k}{\delta^3 k} = \frac{k^2 \Delta k}{\pi^2} V = \frac{8\pi\nu^2}{c^3} \Delta\nu V \qquad (4)$$

modes. The extra-factor 2 accounts for the two possible polarizations for each k vector. 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

$$\frac{dW}{d\nu} = \frac{dM}{d\nu} \cdot h\nu \cdot \bar{n}_1(\nu) = \frac{8\pi\nu^2}{c^3} V h\nu \frac{1}{e^{h\nu/kT} - 1} \qquad (5)$$

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

is the average photon number for <u>each</u> mode whose k vector lies on the spherical surface of radius k .
The distinction between <u>spontaneous</u> and <u>stimulated</u> emission came in the 1917 Einstein's derivation of Eq. (6), as follows. Consider two relevant levels of an atom separated by the energy $\Delta E = h\nu$ and coupled by an optical transition. Each time the atom goes up (<u>absorption</u>) or down (<u>emission</u>), this is a one-photon exchange process. (fig. 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 ν).

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

$$N_1/N_2 = e^{\Delta E/kT} \qquad (6)$$

and equating the rates of absorption and emission

$$N_1 B' \bar{n} = N_2 B \bar{n} + N_2 A \qquad (7)$$

From this latter equation

$$\bar{n} = \frac{A/B}{\frac{B'}{B}\frac{N_1}{N_2} - 1} = \bar{n}_1(\nu)\,\Delta\nu .$$

By use of Eq. (6) and comparison with (5), we get

$$\frac{A}{B} = \frac{8\pi\nu^2}{c^3} V\,\Delta\nu = \Delta M \qquad (8)$$

$$B' = B . \qquad (9)$$

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. 3.

With the probabilities there indicated, the stimulated emission probability into the mode with n photons is larger than the total spontaneous emission over the empty ΔM modes (all those within the linewidth of the atomic emission) when

$$n > \Delta M. \qquad (10)$$

Let us call ΔN the atomic population difference between upper and lower states, P the rate of excitation (pump), n the photon number in the laser mode and

$$T_c = \frac{L}{c}\frac{1}{\theta} \qquad (11)$$

the decay time of photons in the cavity made of two facing mirrors separated by a length L . T_c is equal to $1/\theta$ transit times, since the limited mirror transmittivity $\theta = 1-\rho < 1$ increases the number of transits. Condition (11) stems from considering photons as particles. It is a necessary, but not sufficient condition. Indeed, if we account for wave propagation and phase matching between forward and backward waves, the cavity is resonant for those frequencies corresponding to the standing wave condition (fig. 7)

$$m\ \lambda/2 = L \qquad (m \text{ integer})$$

which amounts to a minimum frequency separation

$$\Delta\nu_{m,m+1} = c/2L \qquad (12)$$

Only for these resonances the escape time is given by (11), otherwise it is much faster (just one transit time L/c).

The rate equations for photons and a population inversion are then

$$\frac{dn}{dt} = B \cdot \Delta N n - n/T_c$$
$$\frac{d\Delta N}{dt} = P - B \cdot \Delta N n \qquad (13)$$

where we have neglected spontaneous processes.
Solving them at equilibrium, the first gives

$$\Delta N = \frac{1}{B T_c} \qquad (14)$$

and the second

$$P = B \cdot \Delta N n \qquad (14)'$$

Combining the two with (10), the pump rate must be

$$P > \Delta M / T_c \quad . \qquad (15)$$

Let us now introduce the concept of atomic cross section σ per atom. The stimulated emission rate Bn can be written as

$$B n = \sigma \Phi \qquad (16)$$

where $\Phi = c n / V$ is the photon flux and hence

$$\sigma = \frac{B V}{c}$$

When the atomic line is broadened only by spontaneous emission process, then, we can put $\Delta \nu = A$ in eq. (4) and have

$$\Delta M = \frac{8\pi}{\lambda^2} \frac{V B}{c} \Delta M$$

Hence

$$\sigma = \frac{\lambda^2}{8\pi} \qquad (17)$$

If there is an extra broadening $\Delta\nu_a > A$ for collision process or other decays, σ reduces as

$$\sigma = \frac{\lambda^2}{8\pi} \frac{A}{\Delta\nu_a} \quad . \tag{17)'}$$

Cross section (17) holds for a bound electron, while for a free electron it is much smaller (fig. 4) since it is given by the square of the classical electron radius $r_0 \sim 10^{-13}$ cm .

On the other hand, writing the volume as V = S.L, condition (14) can be rewritten as

$$P > \theta \frac{S}{\sigma} \Delta\nu \tag{15)'}$$

which shows that the excitation rate is proportional to the ratio between the laser beam cross section S and the atomic cross section. Fig. 4 shows why bound electrons are better than free electrons. However nowadays using high energy ($\sim$ 1 GeV) free electrons in a storage ring one can produce laser action down to $\lambda \sim 1\,\mu m$. Fig. 5 summarizes the different interactions and the spectral regions covered by lasers.

Once n > Δ M is fulfilled, that is, once the privileged mode has enough photons to neglect spontaneous decay channels, we must also take care for the cavity losses, and by (14) request that

$$B\Delta N \geq 1/T_c \quad . \tag{16}$$

This condition is represented in Fig. 6 for two different Δ N. 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 homogeneous and inhomogeneous linewidth. In the former case a monochromatic transition is broadened by circumstances which are equal for all atoms in the cavity (as spontaneous lifetime broadening in a gas, phonon interaction in a solid matrix). All atoms can contribute over the whole 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. 7 two different modes have nodes and maxima in different positions, hence they will "exploit" different atoms, releasing the competition. The simultaneous laser action over many modes is then possible.

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.

Another inhomogeneity occurs in a crystal where active ions are exposed to a crystal field which changes from site to site.

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

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

In Fig. 8 we have shown the scheme of mode locking operation. In that figure, the several parts have the following meaning:

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 transform of the amplitude spectrum a), provided the phases are all equal
c) practical scheme of a many-mode 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 $\tau_{dye} \ll 2$ L/c and then recovered after a transient 2 L/c. This corresponds to having a narrow pulse bouncing forth and back between two mirrors. Notice that, from b) the pulse duration is $1/\Delta\nu_a$.

In the scheme of Fig. 9 we have thus 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

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

hence

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

Using ions of a transition element embedded in a crystal or glass matrix, as Cr^{3+} in Al_2O_3 (ruby), or Nd^{3+} in glass, one may have large $\Delta\nu_a$. 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. 8, and <u>not</u> by electronic shutters.

2. Stimulated emission and nonlinear optics (NLO)

We have seen (fig. 3) that the transition rate for an emission process is B . 1 if spontaneous, or B (n+1) if stimulated.

Also in higher order processes as those studied in NLO we can have a spontaneous and a stimulated version. Take a parametric process implying the annihilation of one quantum $\hbar\omega_1$, and the creation of two quanta $\hbar\cdot\omega_2$ and $\hbar\cdot\omega_3$ as in Fig. 9. The transition rate is B for spontaneous emission in the field 2 or B $(n_2 + 1)$ for stimulated emission in the field 2.

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

$$P_i = \varepsilon_0 \chi^{(1)}_{ij} E_j \tag{17}$$

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). Thses non linearities on the same light beam are described by a nonlinear polarization index as

$$P_i = \chi^{(4)}_{ijk\ell} E_j E_k E_\ell \tag{18}$$

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

$$n = n_0 + n_2 \cdot |E|^2 \tag{19}$$

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 gasses, self actions are due to distortion of the electron cloud.

In Table 1 we show some examples of NLO processes.

T A B L E 1

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

3. Coherence and cooperative phenomena

As shown in Fig. 3, stimulated emission explains mode selection, 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 pulses). But all this has very little to do with coherence.

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, that is, peaked at the minimum energy. 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. 10).

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. 11 which gives a cubic polarization

$$P = G_o E - \beta |E|^2 E \tag{20}$$

and hence a quartic free energy

$$W = - P. E = - \frac{G_o}{2} |E|^2 + \frac{\beta}{4} |E|^4 \tag{21}$$

As E increases, the quartic potential well becomes steeper and steeper (Fig. 12), so that a useful E_o can be reached with a little amount of spread, or statistical fluctuations, around it.

We call coherent this highly excited field state without noise. The field can be described with very good approximation by a complex 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 coherent nonlinear spectroscopy which sheds information on fine properties of atoms and molecules.

4. Practical limitations to coherence

The above picture however is misleading if we aim to long distance interferometry. In fact high order (nonlinear) correc-

tions regard the photon number, that is, the square of the field amplitude and not its phase. Even when the laser amplitude is stabilized by the above nonlinearities, there is a residual phase noise giving an ultimate linewidth as (Townes formula)

$$\Delta\nu = \frac{\pi h \nu}{2P} (\Delta\nu_c)^2$$

where $\Delta\nu_c$ is the cavity width (that is, the resolution of the empty laser interferometer) and P the power output. For $P \sim 1$ mW it would yield $\Delta\nu << 10^{-2}$ Hz.

A more stringent limitation is imposed by practical features as the thermal noise on the cavity or mechanical fluctuations in the laboratory room. Practically, today one can achieve a long term stability around 10 k Hz.

Figure Captions

1 - Spherical shell in K-space.

2 - Radiative transitions in a two-level atom.

3 - Decay channels of an excited atom into different field modes.

4 - Radiative electron cross-section versus frequency.

5 - Map of coherent radiation emission mechanisms.

6 - Scheme of two standing waves in a Fabry-Perot cavity, and interplay between cavity resonances and atomic gain line.

7 - Intensity distributions of two standing waves.

8 - Mode locking operation.

9b- Angular relation in a non-linear optical process.

9a- Non-linear process.

10 - Harmonic potential well and equilibrium statistical distribution for a linear field.

11 - Third-order radiative process in a two-level atom.

12 - Quartic potential well and statistical distribution for a laser field above threshold.

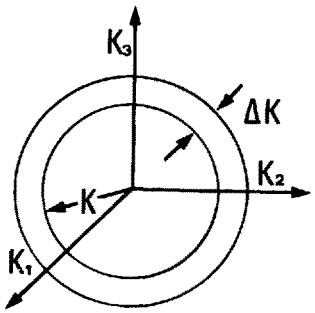


Fig. 1

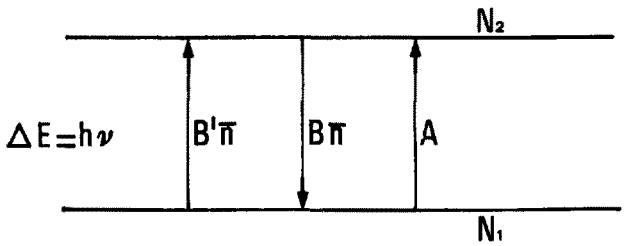


Fig. 2

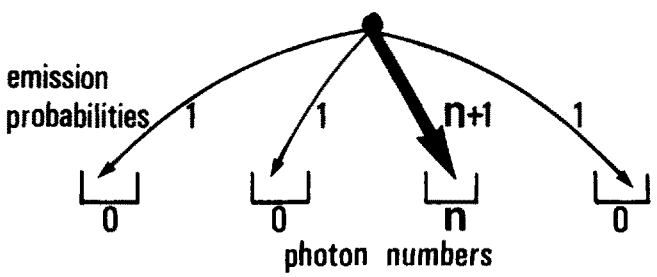


Fig. 3

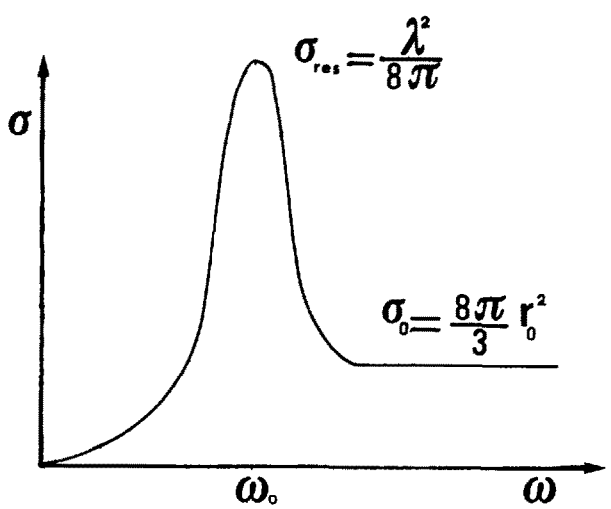
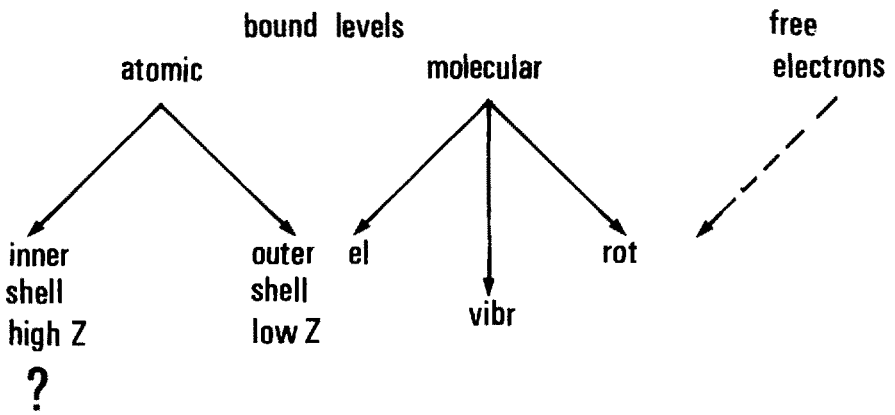


Fig. 4

LASERS


bound levels
atomic
molecular
free
electrons
inner
shell
high Z
?
outer
shell
low Z
el
vibr
rot

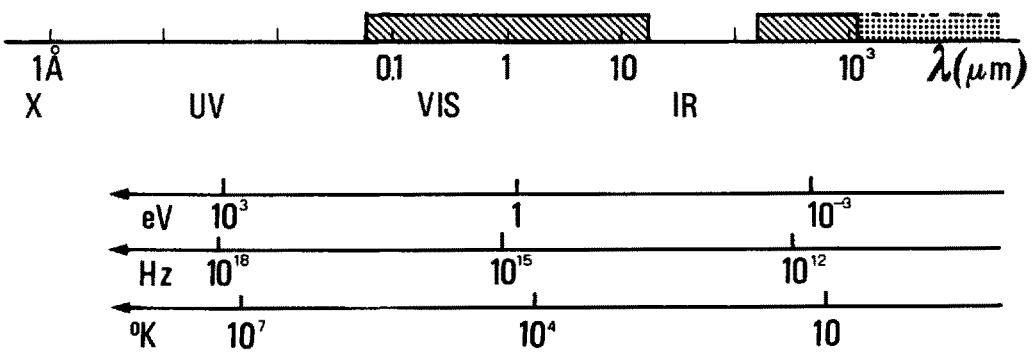
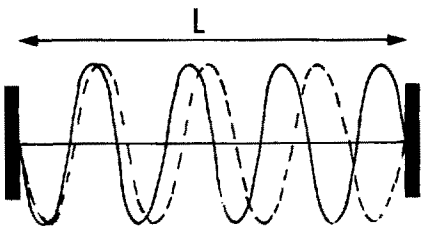

1Å
X
UV
0.1
1
10
VIS
IR
10^3
λ(μm)
eV 10^3 1 10^{-3}
Hz 10^{18} 10^{15} 10^{12}
°K 10^7 10^4 10

Fig. 5



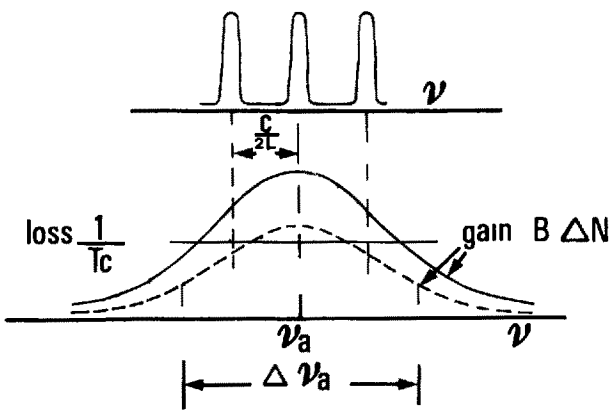
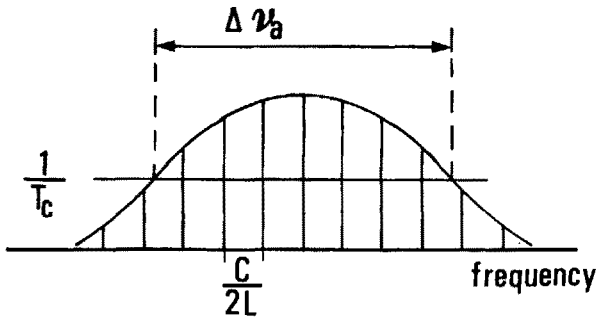
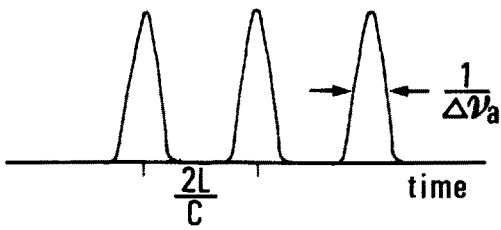


Fig. 6

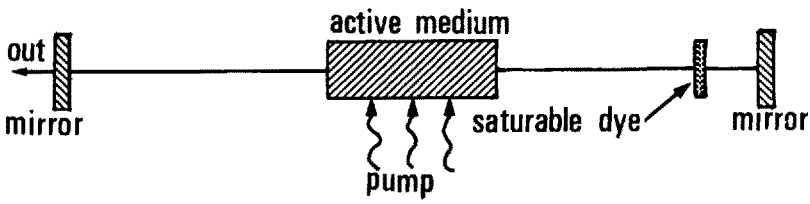
Fig. 7


Δ νa
1/Tc
C/2L
frequency

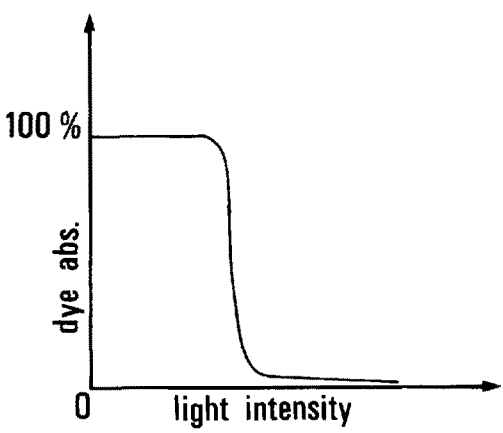
a)


1/Δνa
2L/C
time

b)


out
mirror
active medium
pump
saturable dye
mirror

c)


100 %
dye abs.
0
light intensity

d)

Fig. 8

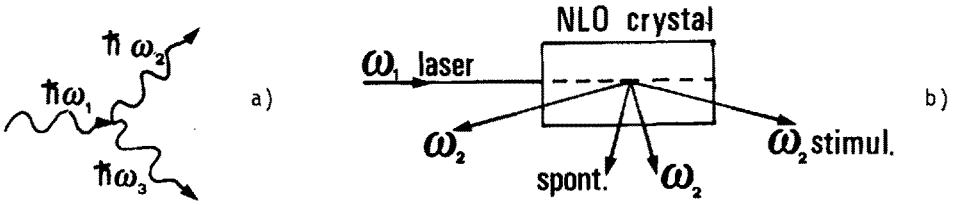


Fig. 9

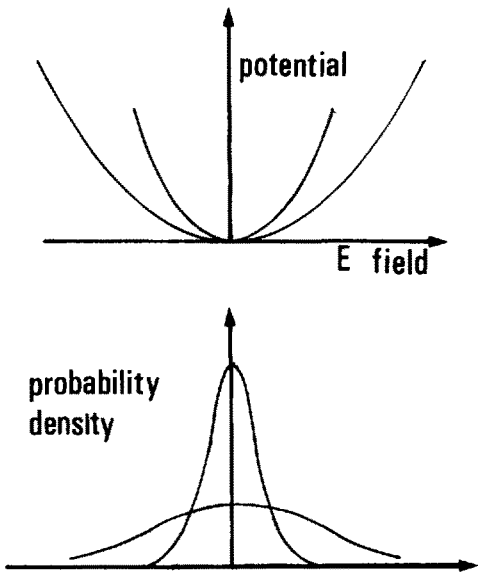


Fig. 10

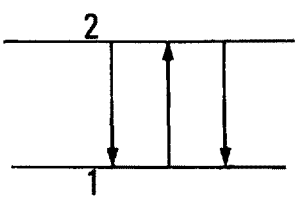


Fig. 11

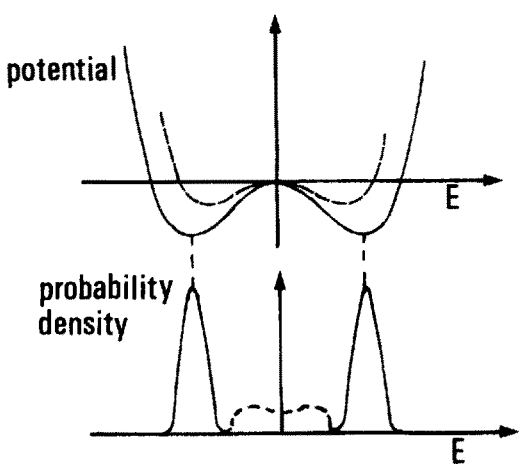


Fig. 12