"Enrico Fermi" Chair
2021/2022

Lectures at Sapienza Università di Roma,
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A History of the Science of Light

From Galileo's telescope to the laser and the quantum information technologies

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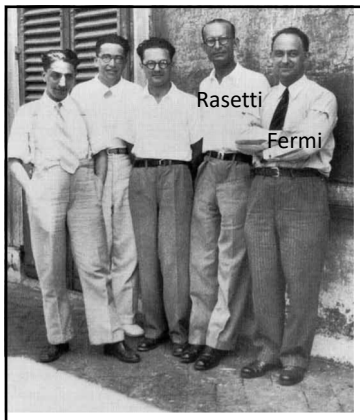
Lecture 9. March 24th 2022

Lecture 9:
Optical pumping methods and the birth of the laser

In the years 1920 to 1950, microwaves and visible light were mainly used to gain information on atoms and molecules by spectroscopy: position of energy levels, magnetic and electric spectral line perturbations were studied (Zeeman, Stark and vacuum fluctuations induced shifts). Then, starting in the 1950's, novel methods using light in order to manipulate atoms and to prepare them in non equilibrium states were developped. Double resonance and optical pumping experiments exploited the exchange of angular momentum between light and matter to orient nuclear or electronic magnetism in gases, providing important information about the interactions between atoms or molecules and their environment. These manipulations led to the invention of the laser which produces light with characteristis very different from the classical light of ordinary lamps. The directivity, monochromaticity and spatio-temporal coherence of laser light has made possible new ways to manipulate atoms and to achieve unprecedented precision in spectroscopy. Laser light also concentrates huge amount of energy in ultra intense and ultra short light pulses, opening the way to the new fields of non-linear optics and extreme light physics. I start to describe this physics in this lecture. After reviewing optical pumping methods, I present a simple model of the laser, analyse the properties of the light it emits, present orders of magnitudes of achievable light intensities and pulse light durations and discuss the new physics now opened to research.



A precursor paper of double resonance by Fermi and Rasetti (1925)

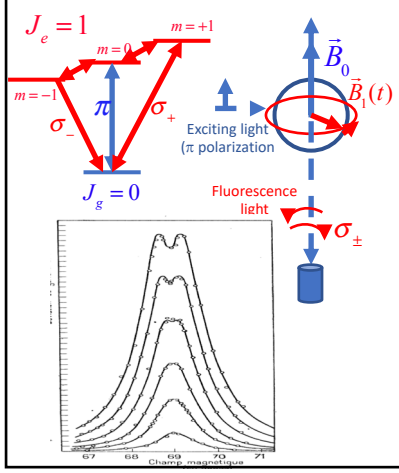
EFFETTO DI UN CAMPO MAGNETICO ALTERNATO SOPRA LA POLARIZZAZIONE DELLA LUCE DI RISONANZA(*)

E. FERMI e F. RASETTI

« Rend. Lincei », *1*, 716–722 (1925 (**).

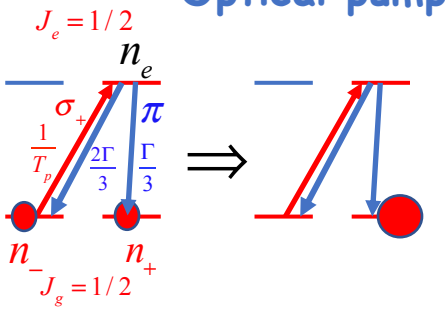
Double resonance (Kastler-Brossel, 1950)

The first precise manipulations of atoms with light beams started before the laser era, in the 1950's, using the broad-band light of spectral lamps. The double-resonance method of Kastler and Brossel involved the preparation of atomic excited states in well defined magnetic substates and the study of magnetic resonance transitions between these states by optical methods. I illustrate this on a simple example: the excitation of a transition between a level of zero angular momentum in its ground state (J_g=0) and a level of angular momentum 1 in the excited state (J_e=1). The atom is placed in static magnetic field B_0 aligned along Oz which splits the excited state into three levels of energies $m\hbar\omega_0$ (m=+1,0 and -1).



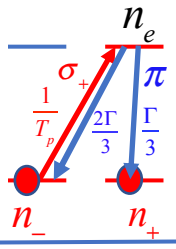
A light beam polarized along Oz (so called π polarization) can excite the J_g=0 towards the J_e=1, m=0 state while a σ_+ circularly polarized beam can excite the transition from the ground state towards the J_e=1, m=+1 sublevel (conservation of angular momentum). If the atom is excited by a π polarized light beam, it will subsequently fall back to the ground state by spontaneous emission, emitting a π polarized photon. Suppose now that the atom is simultaneously submitted to a radiofrequency field resonant at frequency $\omega=\omega_0$ connecting the m=0 to the m=1 sublevel of the J_e=1 excited state. In steady state operation a fraction of the order of ω_1^2/Γ^2 of the atoms is transferred towards the m=1 substate from which they spontaneously emit circularly polarized photons. The intensity of the circularly polarized fluorescence light versus the frequency of the rf field exhibits a resonance around $\omega = \omega_0$. This is a magnetic resonance experiment in which light plays a double role: it prepares the atoms in a state out of thermal equilibrium and detects the resonance with a very high sensitivity. The shape of the resonance is more complex than for a two-level system, since the rf field can also bring the atom into the m =-1 sublevel. The rotating spin is now a spin 1 instead of the spin ½ we have studied earlier. This induces a modification of the resonance line shape (« *Majorana resonance* » profile presenting a dip at line center).

Optical pumping (Kastler-1950)



Optical pumping uses broadband light to prepare the ground state of atoms in a state out of thermal equilibrium and to detect the evolution of the induced atomic polarization by optical methods. The light beam serves at the same time as the preparer and the detector of the atomic orientation. We discuss it in the case of an optical transition linking two F=1/2 levels. The ground state may then be a nuclear spin I=1/2 in an electronic configuration in which the orbital angular momentum is L=0 (S=0 state). This is the simple case of the optical pumping of the Hg^{199} atom. Here again, the conservation of angular momentum plays an essential role. The light is circularly polarized and can only induce the absorption from the m=-1/2 sublevel in the ground state (the number of atoms in this state being called n_-). The atoms are brought by the light in the upper state with m=+1/2 (the number of atoms in this state is n_e). The excited state decays by spontaneous emission with a global rate Γ, 2/3 of the time toward the ground state m=-1/2, 1/3 of the time toward the state m=+1/2 (the number of atoms in this state is n_+). We call $1/T_p$ the pumping rate, proportional to the *absorption* Einstein coefficient and to the intensity of the pumping light beam. We assume (which is generally the case experimentally) that $1/T_p$ is very small compared to Γ ($\Gamma T_p \ll 1$). The cycles of circularly polarized light absorption followed by spontaneous emission act as a « leaky pump » progressively increasing n_+ at the expense of n_-. The pumping process leaks because it succeeds only 1/3 of the time, while 2/3 of the time the atom falls back into the initial state. After a few cycles however, all the atoms accumulate in the m=+1/2 ground state. Starting from thermal equilibrium ($n_-=n_+$), the atomic sample ends up completely polarized ($n_+=n$, total number of atoms, $n_-=0$). We analyze quantitatively the process on next slide.

The optical pumping process



$$\frac{dn_e}{dt} = \frac{1}{T_p} n_- - \Gamma n_e$$

$$\frac{dn_-}{dt} = -\frac{1}{T_p} n_- + \frac{2\Gamma}{3} n_e$$

$$\frac{dn_+}{dt} = \frac{\Gamma}{3} n_e$$

The evolution of the populations of the three states involved in the process is described by Bloch equations depending upon the $1/T_p$ and Γ rates, which translates in mathematical terms the qualitative description of the previous slide. Solving these linear differential equations is simple. It is even simplified further by remarking that n_e remains very small compared to the ground state population throughout the process.

The atoms in the excited state are indeed pumped much more slowly than they are decaying. It is thus possible to eliminate n_e and to get simple rate equations linking n_+ and n_- with the condition $n_+ + n_- = n$. Eliminating n_e between the second and the third equation, we get: :

$$\frac{dn_-}{dt} = -\frac{1}{T_p} n_- + 2\frac{dn_+}{dt}$$

Neglecting n_e compared to n_+ and n_- means that the rates of evolution of n_+ and n_- are nearly opposite:

$$\frac{dn_-}{dt} \approx -\frac{dn_+}{dt}$$

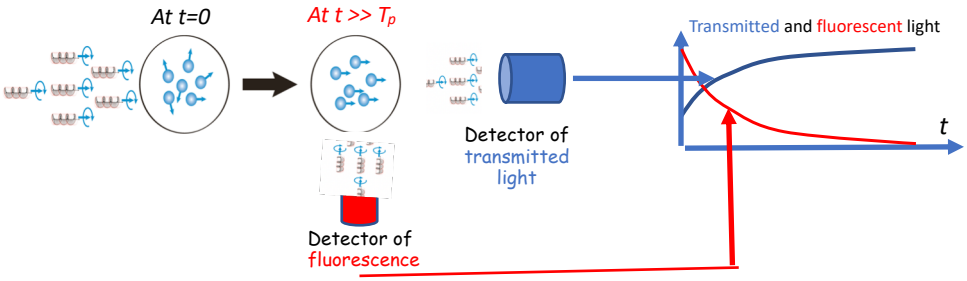
Hence: $$\frac{dn_+}{dt} = -\frac{dn_-}{dt} = \frac{1}{3T_p} n_-$$

and by integrating and taking into account the boundary condition $n_+(t=0)=n_-(t=0)=n/2$:

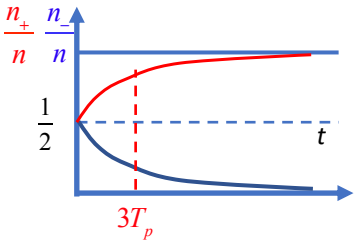
$$\frac{n_-}{n} = \frac{e^{-t/3T_p}}{2}; \frac{n_+}{n} = 1 - \frac{e^{-t/3T_p}}{2} \rightarrow \frac{n_+ - n_-}{n} = 1 - e^{-t/3T_p}$$

Within characteristic time $3T_p$ light transfers its angular momentum to atoms all pumped into m=+1/2 sublevel. (I have neglected here causes of relaxation other than those due to optical pumping and spontaneous emission)

Optical detection of atomic orientation



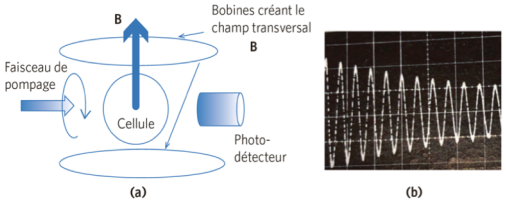
The evolution of the pumping process is detected on the transmitted or fluorescent light



Evolution of m=-1/2 and m=+1/2 state populations

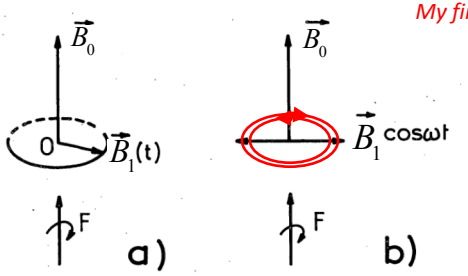
Optical detection of free spin Larmor precession

After equilbrium is reached, the magnetic field is switched to a transverse direction. The transmitted light modulation records the damping of the spins free Larmor precession



Multi-photon transitions in OP experiments with oscillating rf field: an example of angular momentum conservation

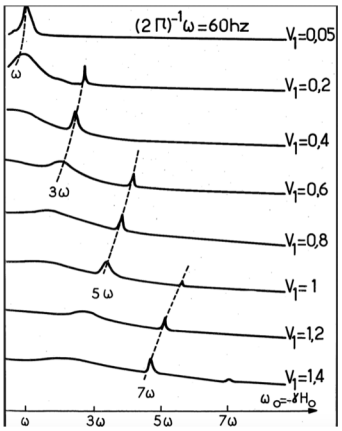
My first experiment (1965)

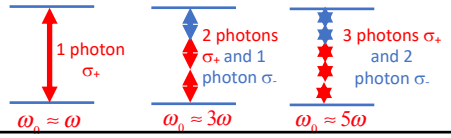


Circularly polarized rf field (time independent evolution in rotating frame: only one resonance for $\omega=\omega_0$)

Linearly polarized transverse rf field is the superposition of σ_+ and σ_- rotating fields: perturbative solution predicts multi-photon resonances

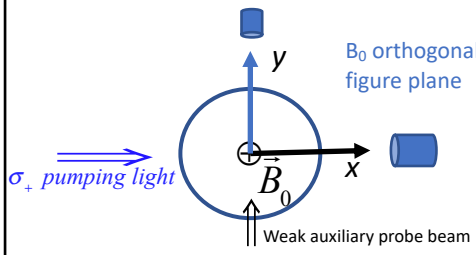
Absorption of n+1 σ_+ photons and n σ_- photons increases by one unit the atom's spin which flips from m=-1/2 to m=+1/2, conserving energy and angular momentum of atom +field system. Even photon number transitions are forbidden with this transverse rf polarization. The resonances are shifted towards small fields by an amount proportional to the rf field intensity B_1^2(« Bloch-Siegert » shifts)





Spectrum of magnetic resonance observed by recording the fluorescent light while sweeping B_0 with a $B_1\cos\omega t$ field at 60Hz: when the spins are flipped, the fluorescence resonantly increases. The usual resonance is observed around $\omega=\omega_0$ and a spectrum of odd-frequency multiphoton resonances are detected for $\omega_0 = 3\omega$, 5ω, 7ω etc... The spectra are recorded, from top to bottom,with increasing B_1 amplitude (measured by V_1, the voltage between the terminals of the rf coils)

Zero field Resonance in transverse pumping: Hanle effect



Optical pumping orients the spins along Ox with a static field B_0 applied along Oz. The transmitted light is recorded while B_0 is swept around $B_0 = 0$. The atomic spins evolve under two conflicting influences: the pumping tends to maintain them along Ox, while the static field makes them rotate in the Oxy plane. When B_0 is such that the spins fan out over a 2π angle during a time of the order of T_p, the orientation is completely lost: One thus observes in the transmitted light a resonance around $B_0=0$ with a width proportional to $1/T_p$. The resonance shape is a Lorentzian absorption curve for P_x. If one detects instead the light transmitted along Oy by a weak auxiliary probe beam, the resonance shape is a dispersion Lorentzian curve.

In order to derive the Bloch equation describing this situation, we define now the spin polarization along Ox as:

$$P_x = \frac{n_+ - n_-}{2n}$$

The P_x and P_y components of the spin obey the following equations (assuming that OP is the only cause of relaxation):

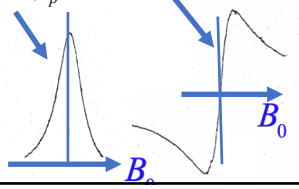
Larmor precession

$$\frac{dP_x}{dt} = -\frac{1}{3T_p}(P_x - 1) - \omega_0 P_y \qquad \frac{dP_y}{dt} = -\frac{1}{6T_p}P_y + \omega_0 P_x$$

Optical pumping and relaxation (P_y relaxes twice slower than P_x)

with the steady state solution:

$$P_x = \frac{\gamma_p^2}{\omega_0^2 + \gamma_p^2} \qquad P_y = \sqrt{2}\frac{\omega_0 \gamma_p}{\omega_0^2 + \gamma_p^2} \qquad \left(\gamma_p = \frac{1}{3\sqrt{2}T_p}\right)$$



This is the Hanle effect, a special case of level-crossing resonance (see Lecture 10)

Different mechanisms of nuclear spins optical pumping

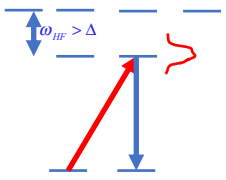
Spectroscopic notation: atomic energy levels are labelled by the value of the principal quantum number in the external orbital n, the total orbital electronic momentum L, the total electronic spin S, the value of the sum J=L+S and, if there is a nuclear spin I, the value of the total angular momentum F=J+I.

$$n^{2S+1}L_{J=L+S};\ F = J + I$$

$L = 0,\ 1,\ 2, 3.....$
S, P, D, F

Nuclear spins Larmor precessions are of the order of 1kHz/Gauss

Nuclear spin orientation via hyperfine coupling (case of Hg^{199})

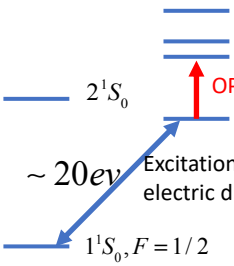


6^3P_1, F=3/2
6^3P_1, F=1/2
6^1S_0, F=1/2

Hyperfine splitting due to interaction of electron with nuclear spin

OP connects only two F=1/2 states, as discussed above. The « naked » nuclear spin I=1/2 is not directly sensitive to the light's electric field. The transfer of angular momentum from light to nuclear spin occurs via the hyperfine coupling in the excited state: during the correlation time $1/\Delta$ of the atom-light interaction and during the life time $1/\Gamma$ of the excited state, the nuclear spin efficiently couples to the electronic variables sensitive to light.

Nuclear spin orientation via hyperfine coupling and metastability exchange (case of He^3)



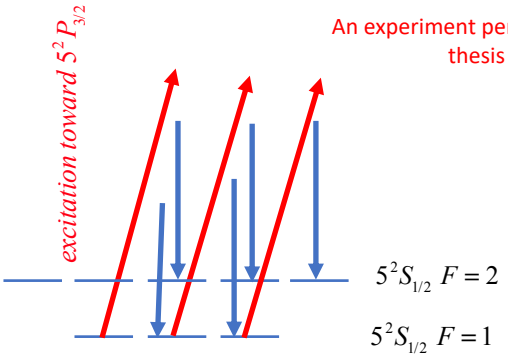
The long lived metastable state 2^3S_1 populated by electric discharge is pumped by infrared light and the nuclear spin oriented via the hyperfine coupling as for ^{199}Hg.

Metastability exchange collision between atom A in 2^3S_1 and atom B in 1^1S_0:

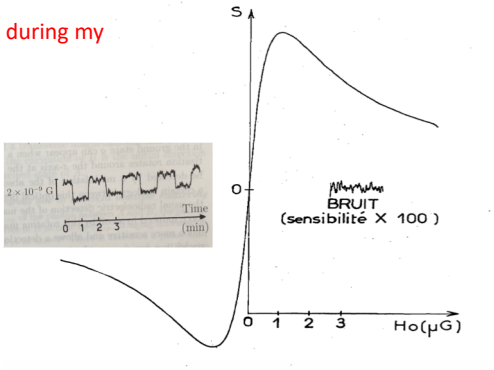
$$(A: 2^3S_1) + (B: 1^1S_0) \rightarrow (A: 1^1S_0) + (B: 2^3S_1)$$

During the very short collision time, the nuclear spin remains unaffected and the ground state is efficiently oriented.

Rubidium atom optical pumping magnetometer



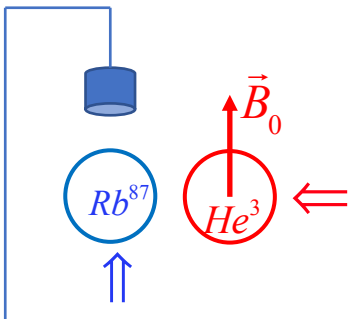
An experiment performed during my thesis (1969)

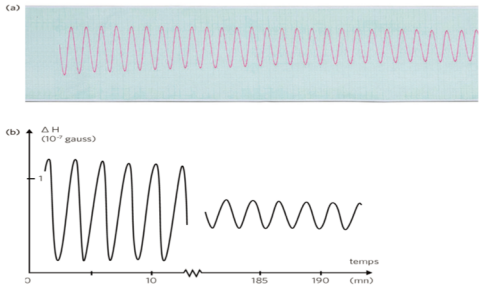


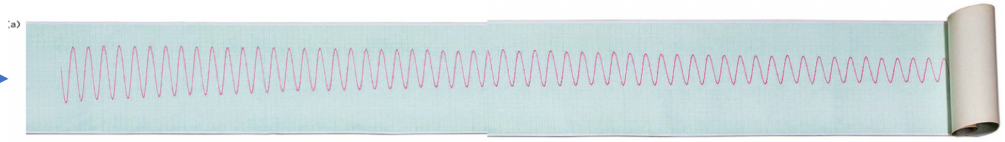
Pumping with circularly polarized light inverts populations in hyperfine ground state (empties the F=1 state) and orients the angular momentum in the F=2 state: the energy and angular momentum of the atom + light system is conserved. A 100 cm3 cell of optically pumped Rb^{87} realizes a very sensitive magnetometer.

Dispersive zero-field Hanle resonance of ground state Rubidium atoms: the atoms are pumped along Ox and a small magnetic field is swept around 0 in the Oz direction. The electronic magnetism corresponds to a Larmor frequency three orders of magnitude larger than that of nuclear magnetism. The relaxation time is about 1s and the line-width of the order of 1 μG, with a signal to noise ratio of the order of 10000: the Rb magnetometer is sensitive to variations of the magnetic field of the order of 10^{-10} Gauss detected in a few seconds (experiment made in a mu-metal magnetic shield).

A double optical pumping experiment: a Rb magnetometer detects the Larmor precession of He^3 nuclear spins



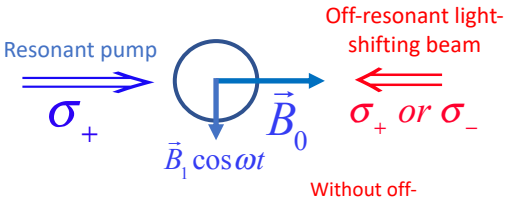


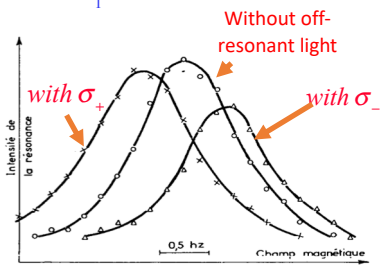


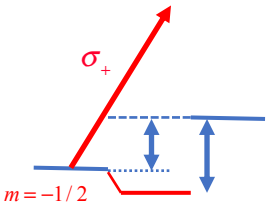
Slow Larmor precession (one period in 90 s) observed for more than ten hours: demonstrates the high sensitivity of the Rb magnetometer and the long life-time of the He^3 nuclear orientation (used since then for medical applications)

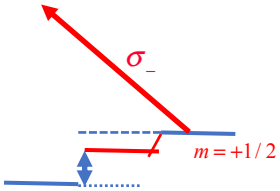
Discovery of light shifts (Cohen-Tannoudji, 1961)

A non resonant light shifts the energy levels by an amount proportional to the light intensity. If the light frequency is close to an atomic transition, the shift is inversely proportional to the frequency mismatch and its sign depends upon the sign of the mismatch (blue or red detuning). We will discuss these shifts later in more details. Here we describe how they have first been observed in an optical pumping experiment of Hg^{199} by C. Cohen-Tannoudji in 1961.





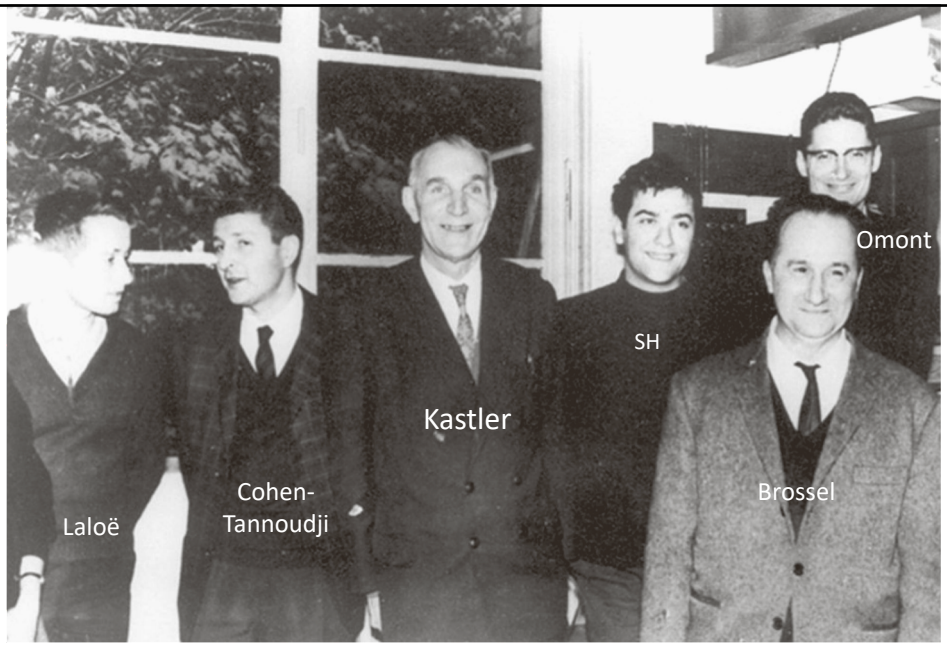




Off-resonant light σ_+ shifts the m=-1/2 state downwards and increases the magnetic resonance frequency

Off-resonant light σ_- shifts the m= +1/2 state downwards and decreases the magnetic resonance frequency

The off resonant light was produced by a lamp containing a different isotope of mercury. The shift was only about 1Hz. Light shifts billions of times larger are currently produced with laser light today (see lecture 11)



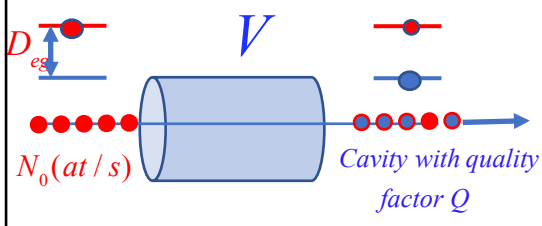
Alfred Kastler Nobel Prize announcement (October 1966)

The Laser and the beginnings of non-linear optics

The development of magnetic resonance experiments has opened new perspectives in the field of light-matter interaction studies. The possibility to prepare matter out of thermal equilibrium, first with Stern-Gerlach devices, then in optically pumped gaseous samples, was promising to realize the amplification of radiation that Einstein had predicted in 1916. Magnetic resonance experiments have also shown that one could observe non-linear effects in matter-radiation interaction (saturation of the atomic response, multiple photon transitions…). This new domain blossomed with the invention of the Maser, then the Laser. These devices are based on fundamental non-linear properties of matter-radiation interaction which I will briefly review in this lecture, before describing next week how they have revolutionised the field of atomic and molecular spectroscopy.

I will first recall the principle of the maser-laser operation, then briefly introduce the main properties of this new source of radiation whose performances have been steadily increased ove the last sixty years.

Simple model of Maser



A beam of atoms or molecules prepared in the upper state of an electric dipole transition (of matrix element D_{eg}) by a Stern- Gerlach or an optical pumping scheme cross an electromagnetic cavity tuned to be resonant with the field radiated by the beam. The flux of entering particles is N_0 per second, the cavity volume is V and its quality factor measuring its losses is Q. The atoms undergo a Rabi oscillation from e to g, each particle delivering a photon of angular frequency ω.

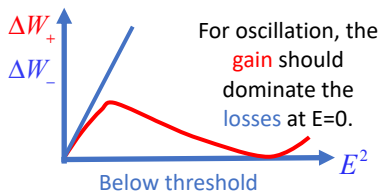
Assume for simplicity that the field is spatially uniform in the cavity with an amplitude E and that all atoms, having the same velocity, cross the cavity in a time of flight T_0.

The energy brought per second by the flux of atoms or molecules to the cavity field is:

$$\Delta_+ W = N_0 \hbar\omega \times \sin^2 \frac{D_{eg} E T_0}{2\hbar}$$

And the energy lost per second by field damping is:

$$\Delta_- W = \frac{\omega}{Q} \varepsilon_0 E^2 V$$



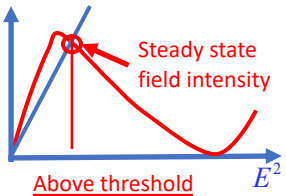
For oscillation, the gain should dominate the losses at E=0.

Below threshold

Condition for self-oscillation:

$$\frac{N_0 \left| D_{eg} \right|^2 T_0^2}{4\hbar\varepsilon_0 V} \geq \frac{1}{Q}$$

The maser operating point depends on the non-linearity of the atomic response.



Above threshold

typical parameters

$(V = 10^{-3} m^3, T_0 = 10^{-3} s,$
$Q = 10^5, \left| D_{eg} \right| \sim q_e a_0)$

$N_0 \geq 4.10^{11} atoms/s$

Simple model of Laser: reminder about linear optics

In order to get a simple model of a laser (optical maser), we must take into account the spatial variation of the light field in the cavity. Let us write Maxwell's equation in a medium filled with polarizable particles. The Maxwell-Ampère equation we have written in Lecture 2 in vacuum must take into account the microscopic currents generated by the bound charges in the atoms or molecules driven by the light field. The density of atomic dipoles P is equal to the number of atoms per unit volume N, multiplied by the electron charge q_e and by the microscopic separation r_{bound} between the positive and negative charge within each atom. The current of bound charges is equal to N multplied by the elementary charges and their velocity v =dr_{bound}/dt.

$$\vec{j}_{bound} = Nq_e \frac{d\vec{r}_{bound}}{dt} = \frac{d\vec{P}}{dt}$$

Introducing this expression in the Maxwell-Ampère equation, we get:

$$\vec{\nabla} \times \vec{B} = \mu_0 \left(\frac{\partial \vec{P}}{\partial t} + \varepsilon_0 \frac{\partial \vec{E}}{\partial t} \right)$$

As we did before (lecture 2), we can eliminate B with the help of the Maxwell-Faraday equation and we get, for the propagation along Oz of a plane wave polarized along Ox, the new form of the d'Alembert equation:

$$\frac{\partial^2 E_x}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E_x}{\partial t^2} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2 P_x}{\partial t^2}$$

In linear optics, P is proportional to E, the proportionality constant χ being the complex susceptibility of the medium which is dependent on the angular frequency of the light:

$$P_x(\omega) = \varepsilon_0 \chi(\omega) E_x(\omega)$$

Searching a complex plane wave solution: $E_0 e^{i(kz-\omega t)}$

we find the dispersion relation (assuming $|\chi|$<<1):

$$k \sim \frac{\omega}{c}\left[1 - \chi'(\omega)/2 - i\chi''(\omega)/2\right]$$

and: $E = E_0 \exp\left[-i\omega(t - n(\omega)z/c)\right]\exp\left[-z/2l(\omega)\right]$

n (ω) is the index of refraction and *l(ω)* the absorption length:

$$n(\omega) = 1 - \chi'(\omega)/2 \;;\; l(\omega) = -c/\omega\,\chi''(\omega)$$

Simple model of Laser: importance of medium non-linear response

$$E = E_0 \exp\left[-i\omega(t - n(\omega)z/c)\right]\exp\left[-z/2l(\omega)\right] \qquad n(\omega) = 1 - \chi'(\omega)/2 \;;\; l(\omega) = -c/\omega\,\chi''(\omega)$$

The features of the linear response of a polarizable medium to a light field resonant with an atomic or molecular transition confirm the qualitative analysis made in our second lecture: The component of the medium polarization in phase with the impinging field ($\chi'(\omega)$) accounts for the medium index, while the component $\pi/2$ out of phase ($\chi''(\omega)$) is responsible for the energy exchange between the medium and the field.

The medium absorbs light if the atomic system has a positive $\chi''(\omega)$ (the population of the lower state of the transition is then larger than that of the excited state). The medium amplifies light by stimulated emission if $\chi''(\omega)$ is negative, which requires a population inversion.

The regime of linear optics with negative $\chi''(\omega)$ corresponds to an exponential increase of the light beam as it propagates in the medium. At some point, this approximation breaks down because the atomic dipoles saturate and cannot follow the increase of the field. The theory must then take into account the non-linearity of the atomic response. We call I_S the saturating intensity and express the medium polarization as a power expansion in I/I_s:

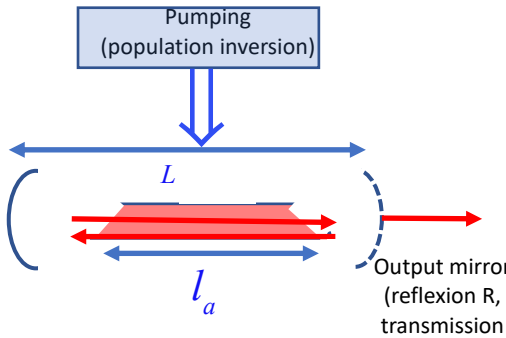
$$\frac{dE_0(z)}{dz} = -\frac{1}{2}\chi''_1(\omega)\left[1 - \frac{I_0}{I_S} + \ldots\right]E_0(z) \quad \rightarrow \quad \frac{dI_0(z)}{dz} = -\frac{I_0(z)}{l(\omega)}\left[1 - \frac{I_0(z)}{I_S} + \ldots\right] \quad ; \quad l(\omega) < 0$$

Simple model of monochromatic laser

The saturation of the amplifying process can be summed up by analogy with the saturation of the magnetic resonance signal, under the compact form:

$$\frac{dI_0(z)}{dz} = \frac{I_0(z)}{l_0}\frac{1}{1+I_0/I_S} \quad ; \quad l_0 = -l(\omega) > 0$$

The amplifying medium (length l_a) is placed in an optical cavity with spherical mirrors separated by distance L. The cavity is tuned to have an integer number of half wavelengths between the mirrors. The ouput mirror is partly transmitting light ouside the cavity which sustains a mode with a transverse Gaussian profile (no need of side walls):



$$\text{Gain in light round trip}: \frac{dI}{I} = \frac{2l_a}{l_0}\frac{1}{1+I/I_S}$$

$$\text{loss in light round trip}: \frac{dI}{I} = 1-R$$

Steady state: Gain = Losses:

$$I = I_S\left[\frac{2l_a}{l_0}\frac{1}{1-R}-1\right] = -I_S\left[\frac{2\omega l_a}{c(1-R)}\chi''_1+1\right]$$

Threshold and steady state regime of laser operation

Threshold condition:

$$\frac{2\omega L}{c(1-R)}\frac{l_a}{L}\chi''_1+1 \le 0 \rightarrow -\chi''_1 \ge -\frac{L}{l_a}\times\frac{c(1-R)}{2\omega L}$$

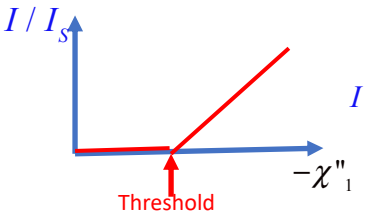
To interpret the right hand side of this equation, let us estimate the damping time of the free cavity field by introducing its dimensionless quality factor Q (number of radians that the field phase accumulates before damping):

$$\frac{dI_{free}}{I_{free}} = -\frac{c}{2L}(1-R) = -\omega\frac{c}{2L\omega}(1-R) = -\frac{\omega}{Q}$$

$$Q = \frac{2\omega L}{c(1-R)} = 4\pi\frac{L}{\lambda}\frac{1}{1-R}$$

We can express the threshold condition in the simple form:

$$-\chi''_1 \ge \frac{L}{l_a}\times\frac{1}{Q}$$



$$I = -I_S\left[Q\frac{l_a}{L}\chi''_1+1\right]$$

« strength » of electric dipole transition

Relaxation time of atomic dipole

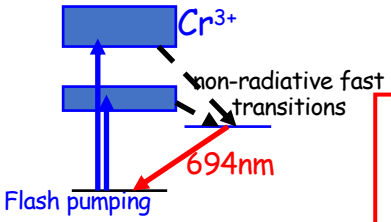
$$-\chi''_1 \sim (n_e-n_g)_{pump}\times\frac{|D_{eg}|^2 T_2}{\hbar\varepsilon_0} \ge \frac{L}{l_a}\times\frac{1}{Q}$$

Population inversion per unit volume

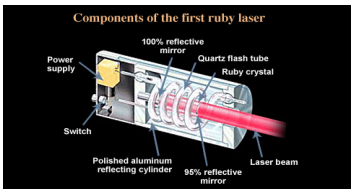
Critical conditions: large population inversion $(n_e\text{-}n_g)_{pump}$ and large cavity quality factor Q

What kind of lasers in the early 1960's?

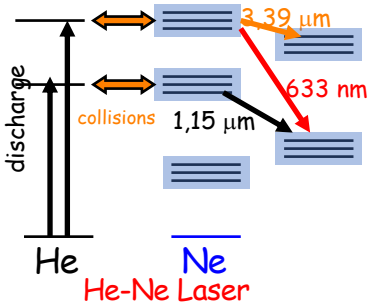
The first lasers had fixed frequencies corresponding to atomic transitions for which an efficient inversion of populations could be achieved



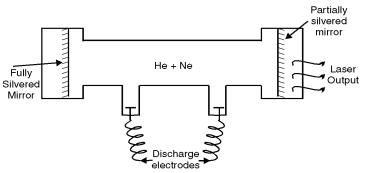
Ruby Laser
(Maiman, 1960)



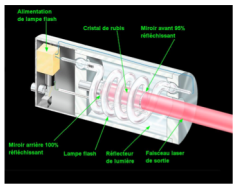
Spectroscopy by coincidence with atomic or molecular lines:
methane transition at 3,39mm (88THz)

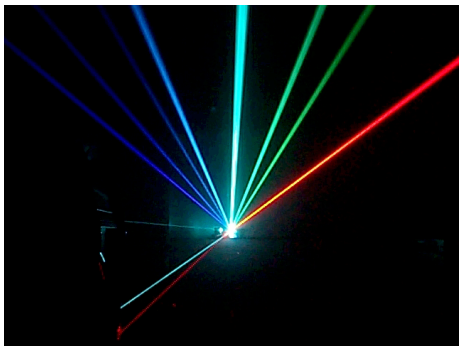


He-Ne Laser
(Javan, 1960)

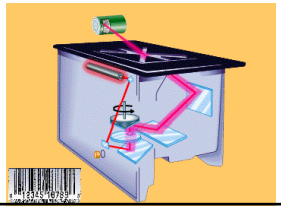


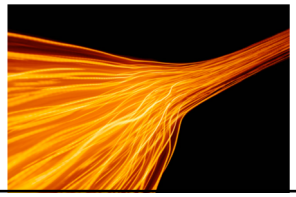
Laser technologies (1960-today)





All kinds of applications.





Fantastic "tamed" light

Intense, directive, monochromatic, coherent….

Fusion and evaporation of matter, cooling and trapping of atoms: lasers can achieve the highest temperatures existing inside stars…and produce the coldest objects in the universe (Bose-Einstein condensates or BEC)

Ultra-stable light beams oscillating without skipping a beat over millions of kilometers…or ultra-short light pulses extending over a few nanometers, crossing matter in a few attoseconds (one billionth of a billionth of a second).

A very flexible tool for fundamental research in physics, chemistry and biology and for applications to metrology, medicine, communication etc…

60 years of Atomic,Molecular & Optical Physics

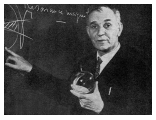
The laser has made tremendous progresses possible and has led to quantitative and qualitative revolutions in basic research

A ten order of magnitude (or more) improvement in many fields (a factor of ten every five years!)

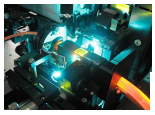
	1960	Today (2022)
Precision (spectroscopy and clocks):	10^{-8} (1s/year)	10^{-19} (<<1s/age of Universe)
Sensitivity of measurements:	10^{10} atoms in cell	1 atom/ 1 photon
Temperatures and kinetic energies of atoms:	1-300K	10^{-10} K (cold atoms)
Speed and time resolution:	Nanosecond (10^{-9}s)	Attosecond (10^{-18}s)
Relative sensitivity to length variation $\Delta h/h$	10^{-8} -10^{-9} (Interferometric definition of meter)	10^{-21} -10^{-22} (LIGO/VIRGO)

Sixty years of revolutions in atomic physics and optics

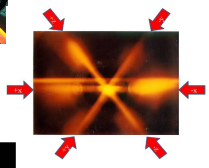
1960/70: Optical pumping, first lasers



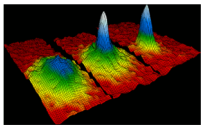
1970/80: Tuneable Lasers and high resolution spectroscopy



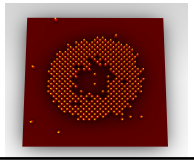
1980/90: Laser cooling, photons in cavities, trapped ions



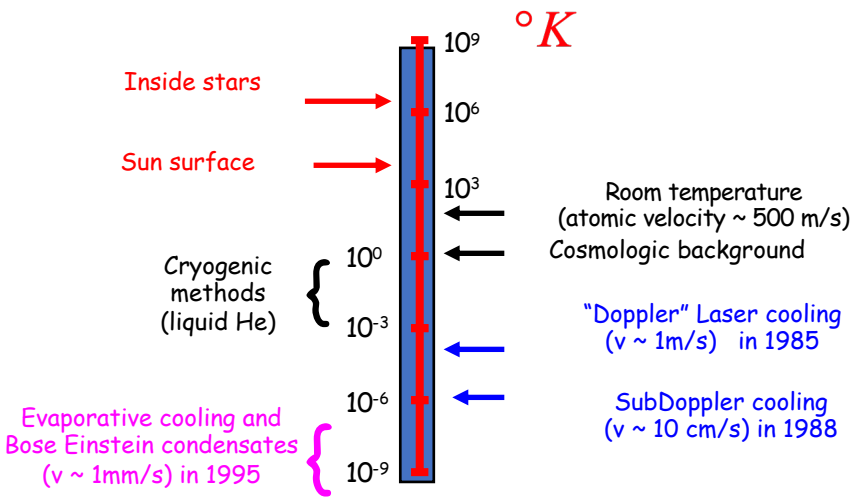
1990/2000: Quantum information, quantum gases, ultra-short laser pulses



2000 - : Manipulation of real and artificial atoms and of light quantum states: atomic physics and quantum optics meets condensed matter physics.



Going towards T=0K



Eleven orders of magnitude gained towards low temperatures and 5 to 6 orders towards low atomic velocities since 1960-70

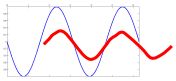
The 1960's: beginning of non-linear optics

P.Franken (1928-1999)

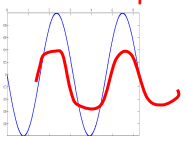
N.Bloembergen (1920-2017)

The high intensity of fixed-frequency lasers made possible the first observations of non-linear optics effects in non-resonant situations or, in case of coincidence beween the frequency of a laser and a transition in a molecular gas, in resonant interactions:

Harmonic generation (non resonant transparent medium):



At low field amplitude E, the atomic dipole D responds linearly



For high field amplitude E, D saturates: the sine wave is flattened and harmonics are generated

In a dense medium, the induced dipoles radiate coherently the harmonic field at frequency multiple of ω (phase matching condition must be fulfilled)

High intensity : D.E ~ $\hbar\omega_{atom}$ (~ 10^{14} W/cm^2)

Symmetry: In atoms with parity symmetry with respect to nucleus, the electric dipole couples only states with opposite parity (even/odd parity states are states whose wave function is multiplied by +1/-1 when changing **r** into **–r**). Due to this symmetry, only *odd harmonics* are generated by atoms. *Even harmonics* are produced by crystals in which atomic environment does not have parity symmetry.

Orders of magnitude of high intensity lasers

Atomic electric field (Coulomb field of proton at Bohr radius distance):

$$E_{atom} = \frac{q_e}{4\pi\varepsilon_0 a_0^2} = 5.1\times10^{11} V/m$$

Light electric field beyond which perturbative development of atomic response in powers of E breaks down:

$$E_{bd} \sim E_{atom}/25$$

Light flux corresponding to perturbative treatment breakdown:

$$P_{bd} \sim \varepsilon_0 c E_{bd}^2 \sim 10^{14} W/cm^2$$

Light flux transforming gas of atoms in a plasma of electrons and ions:

$$P_i = \varepsilon_0 c E_{atom}^2 \sim 7\times10^{16} W/cm^2$$

Ponderomotive energy of charged particle of mass m and charge q in the laser electric field

$$E_{pm} = \frac{1}{2} m \left\langle \dot{z}^2 \right\rangle = \frac{q_e^2 E_0^2}{4m\omega^2}$$

Light flux creating relativistic electrons in a plasma:

$$E^e_{pm} = \frac{q_e^2 E_0^2}{4m\omega^2} = mc^2 \rightarrow E_{er} = \frac{2m\omega c}{q_e} \rightarrow P_{er} = \varepsilon_0 c E_{er}^2 = \frac{4\varepsilon_0 m^2\omega^2 c^3}{q_e^2} \sim 10^{19} W/cm^2$$

Orders of magnitude: still more light power

Light flux accelerating ions with 10^4 electron mass to relativistic energies:

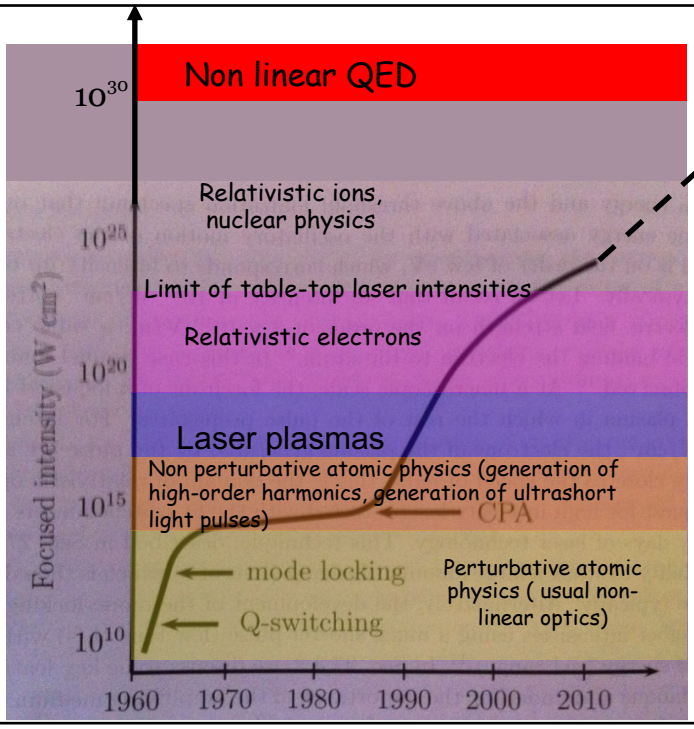
$$P_{ion,r} \sim 10^8 P_{er} \sim 10^{27} W/cm^2$$

Light flux breaking vacuum in electron-positron pairs (non linear QED):

Electric field of light accelerates virtual electron-positron pair during time $\hbar/2mc^2$ permitted by Heisenberg uncertainty relation. Charges propagate over distance $\hbar/2mc$ and acquire an energy $qE\times\hbar/2mc$. When this energy reaches $2mc^2$, the pair can be really created out of vacuum:

$$q_e E_v \times \frac{\hbar}{2mc} \sim 2mc^2 \rightarrow E_v = \frac{4m^2c^2}{\hbar q_e} = E_{er} \times \frac{2mc^2}{\hbar\omega} \sim 10^6 E_{er}$$

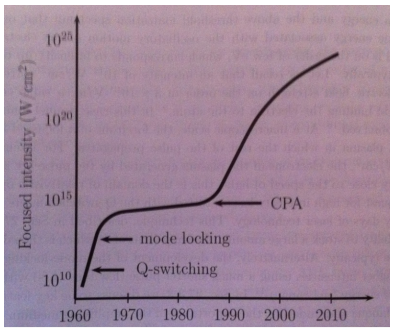
$$P_{vac} \sim 10^{12} P_{er} \sim 10^{31} W/cm^2$$



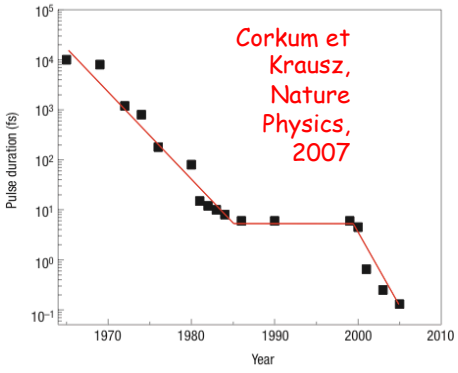
Different domains of intense laser light physics

(Q switching mode locking and CPA explained in subsequent slides)

The increase of laser power has been correlated with the decrease of laser pulse durations



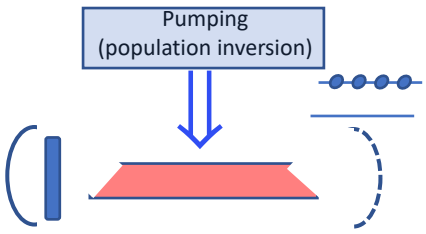
Evolution of laser power (W/cm^2)



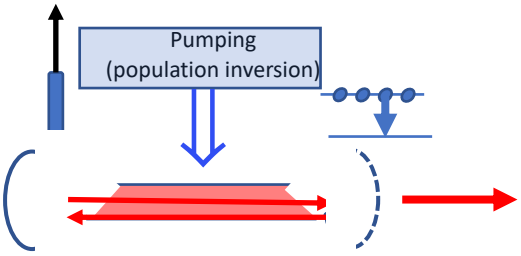
Evolution of pulse duration (femtoseconds)

Two methods to concentrate light energy in short pulses

Q switching:

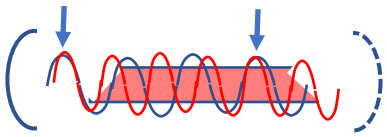


Low Q with a controlable absorber inserted in cavity: the population inversion builds up while the laser remains below threshold

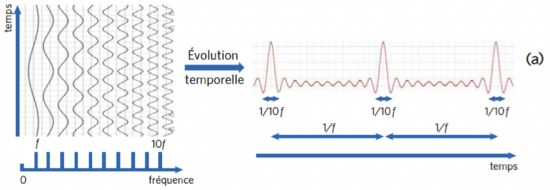


The absorber is removed suddenly, switching the Q to a high value and the laser medium is brought well above threshold: a pulse of light is emitted

Mode locking

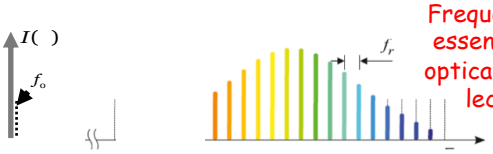


Multimode laser *locked in phase* generate waves which periodically interfere constructively



Example: 10 equally spaced modes produce train of pulses whose width is one tenth of pulse separation

Principle of large band mode locked laser (Ti-Sapphire)



Frequency combs, essential tools in optical clocks (see lecture 12)

If N modes have intensity I_0 with random phases, the laser output is continuous with average intensity:

$$I_m = NI_0$$

If modes are phase-locked, laser emits sequence of pulses with peak intensity N times larger:

$$I_p = N^2 I_0 = NI_m$$

The ratio of the pulse duration τ_p over the pulse repetition period T_p is (conservation of energy):

$$\tau_p / T_p = 1/N$$

Order of magnitude:

$$T_p \sim 2L/c \sim 10^{-8} s \quad ; \quad \tau_p \sim 10^{-14} s$$

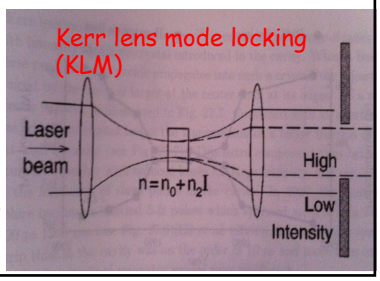
Amplifying medium lasing on a large frequency band Δ with N equally spaced modes separated by frequency interval f = c/2L

$$N = \frac{\Delta}{f} \sim 10^5 - 10^6$$

The locking mechanism is due to non-linear coupling between modes feeding on the same amplifying medium (an effect first observed by Huygens who realized that two clocks suspended on the same wall end-up by beating in phase).

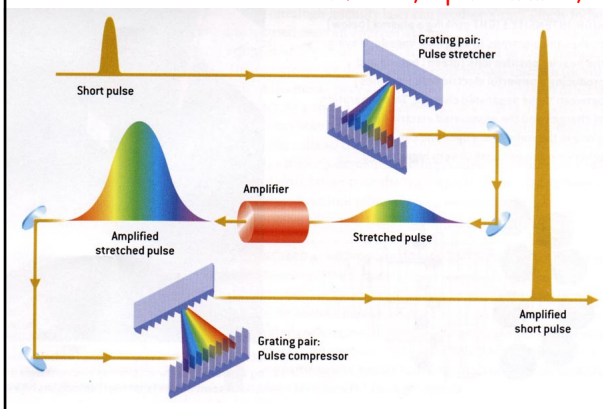
Kerr crystal with intensity dependent index, realize self-focusing of intense light: high intensity is transmitted with less losses, favoring mode locking.

Kerr lens mode locking (KLM)



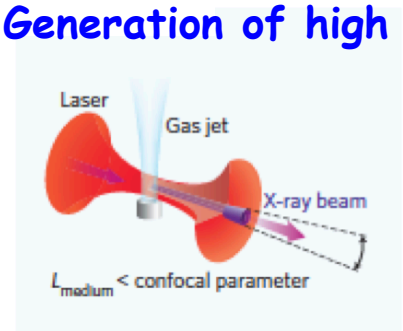
Amplification by pulse stretching and recompressing (CPA method)

Strickland et Mourou, Opt.Comm.56, 219 (1985)



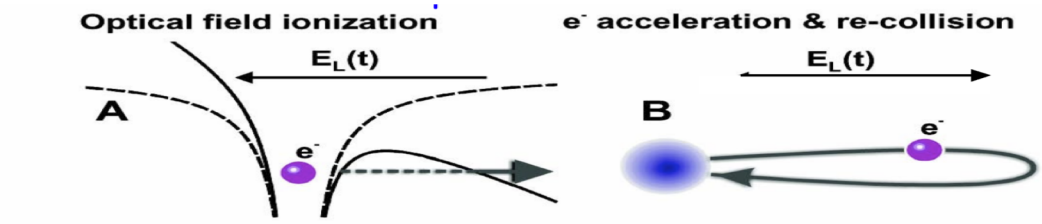
Mode locking restricted until the 1980's the light intensity to a few GW/cm^2 in the amplifying medium, due to saturation and damage produced with higher intensities. This corresponded to a maximum power of ~10^{14} W/cm^2 after focusing of the beam in vacuum. The CPA (Chirped Pulse Amplification) method, has made possible to increase this power by several orders of magnitude. It consists in separating in time the various frequency components of light using gratings, resulting in lengthening the pulses from a few 10^{-15} s to nanoseconds. This decreases the pulse peak intensity, making it possible to amplify them without saturation. A second combination of gratings then recompresses the amplified pulses, realizing a huge gain in intensity. With modern table-top Ti-Sapphire lasers, peak pulse powers of 10^{20}-10^{22} W/cm^2 are achieved, making it possible to reach the regimes of relativistic electron plasma formation. Much higher powers (in the range of 10^{27}-10^{28} W/cm^2 are reached with large laser installations).

Generation of high order harmonics and attosecond x-ray pulses of light



Experiments performed with femtosecond intense light pulses (10^{14} to 10^{17} W/cm^2) exciting beams of rare gas atoms (He, Ne, Xe) . Multiphoton ionization and high harmonic generation is realized. Within a fraction of the optical period, an electron is torn out from the atomic core by the huge electric field of the light beam, then accelerated back towards the core during the next fraction of the optical cycle.

When the electron falls back on the core, the collision produces high harmonics of the laser light, in the far uv and X ray domains. The harmonic pulses have subfemtosecond durations, in the attosecond time domain (1 attosecond= 10^{-18}s). These ultrashort light pulses are employed to study experimentally very fast atomic, molecular and chemical processes in gas and condensed matter.



Concluding remarks

We have started to study in this lecture the methods of atom manipulation with light which have been initiated in 1950 with the double resonance and optical pumping experiments induced by classical lamps. The invention of the laser ten years later has opened a very rich field of research, taking advantage of the remarkable properties of these sources (monochromaticity, spatio-temporal coherence and high intensity). The orders of magnitude of light power fluxes now accessible allow physicists to explore phenomena in which light interacts with matter in very unusual conditions (ultra intense and ultra short light pulses inducing relativistic effects).

In the next two lectures, I will describe experiments exploiting laser light to achieve ultra-high spectroscopic resolution, to cool and trap ions and neutral atoms at extremely low temperatures and to prepare Bose Einstein condensates and quantum degenerate Fermi gases. I will show how these novel systems, born from basic research lead to useful applications.