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I.C.T.P., P.O. BOX 586, 34100 TRIESTE, ITALY, CABLE: CENTRATOM TRIESTE

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MULTIPHOTON IONISATION EXPERIMENTS WITH INTENSE SHORT PULSES

P. Agostini
Service de Physique des Atomes et ds Surfaces
CEN Saclay 91190
Gif Sur Yvette
France

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P. AGOSTINI

Service de Physique des Atomes et des Surfaces
CEN SACLAY 91190 GIF SUR YVETTE

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Generation and measurement of Ultrashort Superintense laser pulses

I. Introduction

During the last two or three years, new laser sources for the production of extremely high peak-brightness outputs have been developped. These new sources are characterized by their small scale, high repetition rate, short pulses and, in some cases, tunability. Peak powers ranging from 0.1 to 10 TWatts are currently achieved with pulse durations from 100 fs to 1 ps, in diffraction limited beams, yielding intensities up to 10^{18} W.cm^{-2}. The prospect for the next few years is to reach 10^{21} Wcm^{-2}. Such sources open new perspectives for research in Atomic Physics, Solid-state Physics, Plasma Physics etc...
The aim of this lecture is mostly to describe the tunable femtosecond laser source which has been used in the experimental investigation of giant Stark shifts induced by intense electromagnetic fields (see lecture II). However, I will begin with some general considerations on intensity and the methods used to measure it. I will pay special attention to time measurements, especially in the case of very short pulses (single-shot autocorrelator). The tunable source consists of a cw, mode-locked oscillator with intra cavity compensation of group-velocity dispersion, subsequently

amplified. The output pulse is used to generate a super-continuum through Self-Phase Modulation in a liquid. SPM is also the basis of another scheme of superintense laser, namely the Chirped Pulse Amplification. Finally, a narrow bandwidth is selected out of the supercontinuum and further amplified to achieve the desired peak intensity.
Time permitting, I will give a quick overview of two other schemes : the multiterawatt Nd-glass laser at Saclay and the ultra high intensity KrF^* system at Chicago.

II. Intensity

It is the incident radiant flux density or flux per unit area ($W m^{-2}$). In the case of a plane wave E, B it is related to the Poynting vector $\vec{S}$ through :

$$I = \bar{S} \qquad (1)$$

where $\vec{S} = \varepsilon_0 c^2 \vec{E} \times \vec{B}$ and $B = \frac{E}{c}$. Hence

$$I = \varepsilon_0 c \overline{E^2} = \varepsilon_0 \frac{c E^2}{2} \qquad (2)$$

where E is the amplitude of the electric field in the electromagnetic wave. In a laser system I is a characteristic of both the laser and the focussing optics. To characterize the source itself it would be preferable to use the "brightness" defined as the radiant flux density per unit of solid angle. This number allows a quick evaluation of various sources : for instance

High Pressure Mercury lamp	250 W/cm²/st
He-Ne Laser	10^6 W/cm²/st
Terawatt laser	10^{22} W/cm²/st

To get an idea of how "intense" a beam is, it is useful to consider the amplitude of the electric field E and to relate it to some physical quantity at the atomic scale. For instance, one may calculate the intensity such that the electric field will be equal to the Coulomb field on the first Bohr orbit:

$$E = \frac{1}{4\pi\varepsilon_0}\frac{q}{a_0^2} = 5.7 \times 10^{11}\ V/m$$

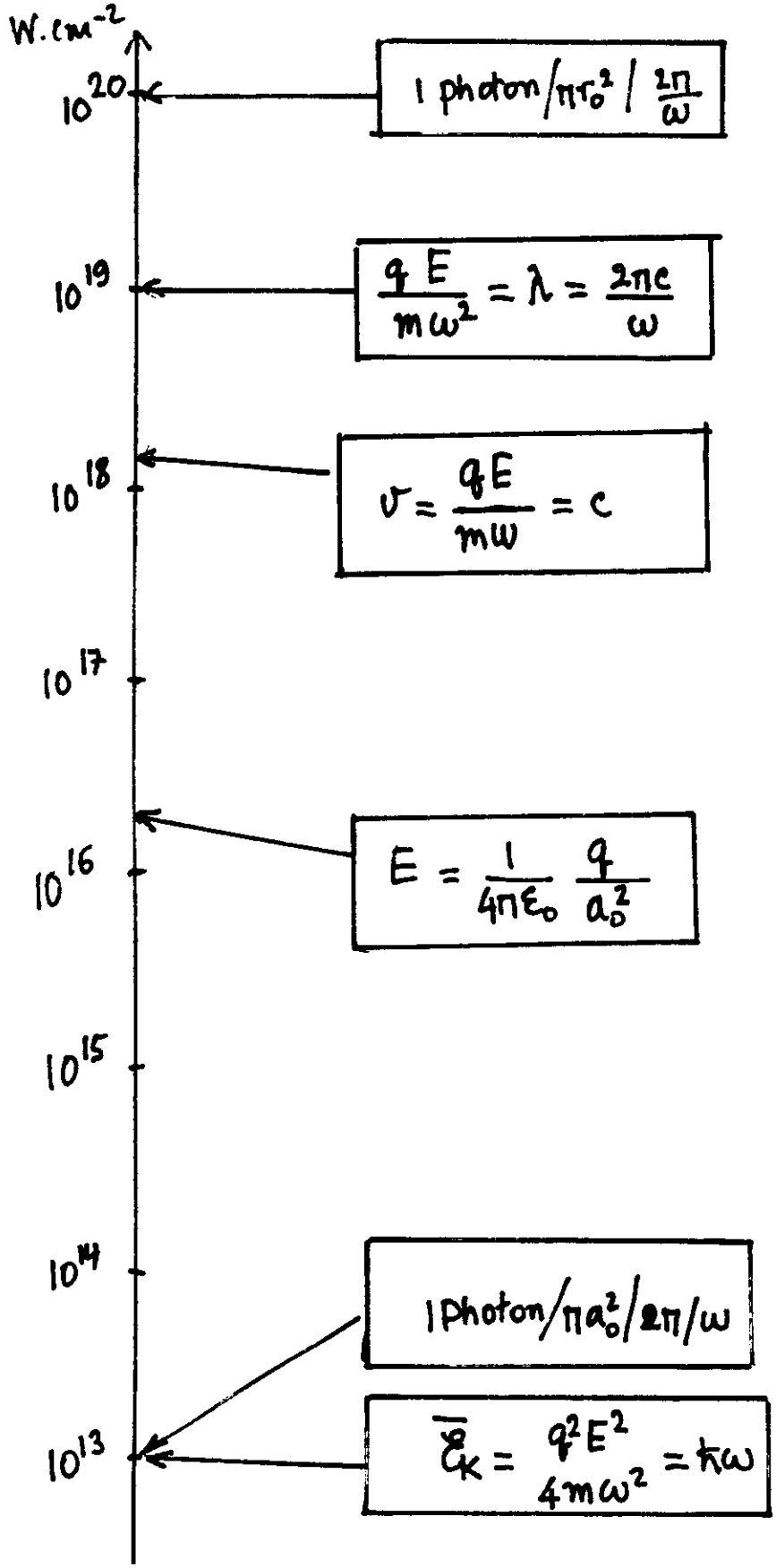
From (2) one gets $I = 4 \times 10^{16}\ W/cm^2$ (atomic unit of intensity). Another effet is particularly well suited to scale the intensity, namely the motion of a free electron forced by the electromagnetic field. This oscillatory (or quiver) motion is related to the so-called "ponderomotive potential" and to the Stark-shifts of Rydberg states induced by the electromagnetic field (see lecture II). This motion is characterized by an amplitude x, a velocity v and a kinetic energy E_K given by, respectively:

$$x = \frac{qE}{m\omega^2} \qquad v = \frac{qE}{m\omega} \qquad E_K = \frac{q^2E^2}{2m^2\omega^2} \qquad (3)$$

Using (2) and (3) it is easy to find that $10^{13}\ W cm^{-2}$ corresponds to an average kinetic energy $\overline{E_K}$ equal to the photon energy $\hbar\omega$ for a wavelength of 1.06 μm (Nd laser). It is also the intensity such that there is 1 photon per field period $\frac{2\pi}{\omega}$ per area πa_0^2 (a_0 : radius of first Bohr orbit).

To force the electron velocity close to c requires an intensity of $10^{18}\ W cm^{-2}$ while having 1 photon per field period crossing

a section πr_0^2, where r_0 is the electron classical radius ($2.8\,10^{-15}$ m), demands 10^{20} W cm^{-2}.



III. Intensity measurements

For a pulsed laser, intensity is redefined as a function of measurable parameters as :

$$I = \frac{W}{S_0 \tau} \qquad (4)$$

where W is the total energy per pulse, S_0 the best-focus section and τ the pulse duration. Given normalized spatial and temporal distributions of intensity $f(x, y, z)$ and $g(t)$, S_0 and τ are defined through :

$$S_0 = \int f(x, y, 0)\, dx\, dy \qquad (5)$$

$$\tau = \int g(t)\, dt \qquad (6)$$

The three quantities W, S_0, τ are separately measured to obtain I.

Energy is measured by a calorimeter or a powermeter usually calibrated by Joule-effect. This measurement does not present particular difficulties.

The beam section S_0 is deduced from photometrics measurements. One technique is, for example, the following : an enlarged image of the focus is formed by a microscope objective on the cathode of a TV camera (or a photodiode array). The image is digitized and analyzed by a computer which calculates numerically quantities (5). The enlargement factor must be calibrated, for instance, using a micrometer.

In the nanosecond range, pulse durations are conveniently measured by recording $g(t)$ by a combination of fast photodiods (response time ~ 100 ps) and large bandwidth oscilloscope ($\sim$ GHz).

In the range of 10 ps or so streak cameras with resolution of one ps can be used. However, for pulse durations of the order of 1 ps and less it is not possible to directly measure $g(t)$ and one must rely on autocorrelation methods.

III.1 single-shot measurement of ultrashort pulses

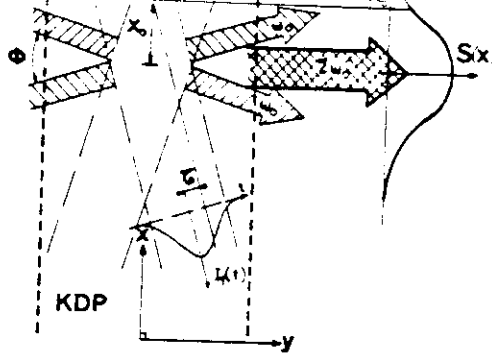
The following method, as described by Salins and coworkers, is currently used.

The basic idea is to transform the temporal shape of the pulse into a spatial shape which could be analyzed by a spatial detector. A beam splitter produces two replicas of the incident pulse which cross each other in a non-linear KDP crystal. The image of the crystal at the second harmonic (SH) wavelength is recorded by a linear array of CCD.

Assume the two incident beams spatially uniform. Let ϕ be the angles between the beams (fig 1) and $I(t)$ their common temporal intensity shape. The second harmonic beam is generated along the symmetry axis (bissector of the two incident beams) due to the cut of the KDP crystal. Inspection of fig. 1 shows that, at a point with abscissa x_0 the instantaneous SH signal is proportional to $I(t-\tau) \times I(t+\tau)$ where

$$\tau = \frac{n x_0 \sin \phi/2}{c} \qquad (7)$$

Fig. 1



where c is the velocity of light and n the index of the crystal. Since the detector integrate the signal over a longer time than the pulse width, the shape $S(x)$ is finally given by:

$$S(x) \propto \int_{-\infty}^{+\infty} I(t+\tau)\, I(t-\tau)\, dt = G_2(2\tau) \qquad (8)$$

All the information is obtained with a single pulse. To calibrate the device it is convenient to introduce a known delay Δt_0 into one of the two beams. The shift of the second harmonic pattern along the x-axis is related to Δt_0 through:

$$\Delta x_0 = \frac{c\,\Delta t_0}{2n \sin \phi/2} \qquad (9)$$

Let's take for example a Gaussian pulse

$$I(t) = e^{-4\,\mathrm{Log}2\,\frac{t^2}{\eta_0^2}} \qquad (10)$$

with a full width at half maximum η_0. The function (8) $G_2(2\tau)$ is easily computed:

$$G(2\tau) = T\sqrt{\pi}\, e^{-2\tau^2/T^2} \qquad \text{with } T = \frac{\eta_0}{2\sqrt{\mathrm{Log}2}}$$

whose full width at half maximum is:

$$\frac{\eta_0}{2\sqrt{\mathrm{Log}2}}\sqrt{2\,\mathrm{Log}2} = \frac{\eta_0}{\sqrt{2}} \qquad (11)$$

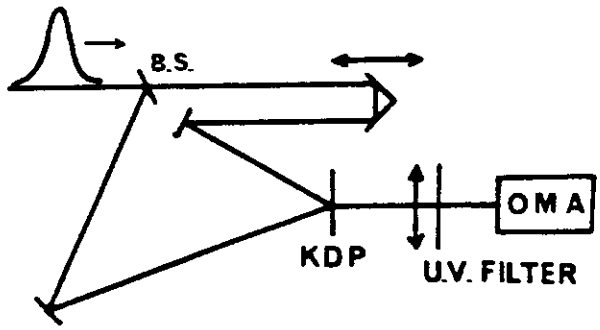
Using (7) and (9) one gets for the FWHM η_0 as a function of the measured FWHM of $S(x)$ δ_0 the expression

$$\eta_0 = \frac{\delta_0}{\sqrt{2}} \frac{\Delta t_0}{\Delta x_0} \qquad (12)$$

In general the factor $\sqrt{2}$ is to be replaced by $1/K$ with $K = 1$ for a square pulse or $K = 1.55$ for a sech^2 pulse.

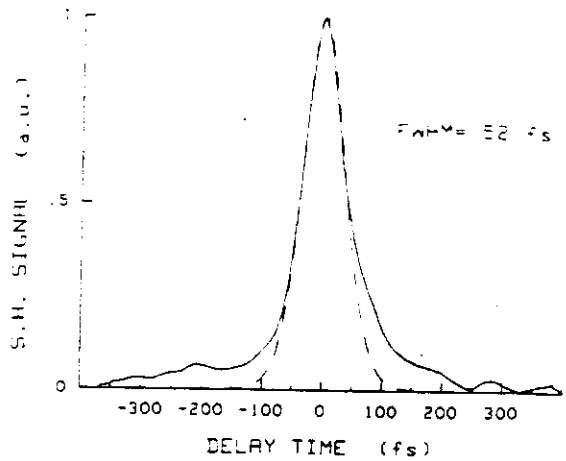
The actual experimental set up is schematized in fig 2.

fig 2



Such a device has proved to be suitable for pulses as short as 50 fs using a 0.3 mm thick KDP crystal, as shown on fig. 3

fig. 3



IV. Tunable Femtosecond laser Source

For a large number of investigations in nonlinear optics and multiphoton processes it is important to have short pulses. One of the reasons is that, the shorter the pulse the higher the intensity actually "seen" by the target. Another reason is the following: if one is interested in the energy spectra of electrons produced by multiphoton ionization, short pulses (< 1ps) are essential to avoid modification of the spectrum due to the ponderomotive force (cf lecture II). Of course, short pulses are essential too for time-resolved studies. Furthermore, in some cases, it is very useful to be able to tune the light frequency.

Several solutions are available to produce tunable, intense picosecond pulses. In the femtosecond regime the most practical source is the supercontinuum generated in liquids by intense femtosecond pulses. The source consists of essentially two parts: the generation of a femtosecond pulse and the generation, filtering and amplification of the supercontinuum.

IV.1 The CPM oscillator with intracavity GVD compensation

As it is well known, the best way to produce short pulses is the mode-locking. By locking in phase all the modes in a cavity, it is possible to generate pulses whose duration is limited by the gain bandwidth of the system. This is easily achieved in a cw-pumped, passively mode-locked

cavity. A practical design is shown in fig. 4. It is a ring-cavity where the amplifying medium is a Rhodamine jet, CW-pumped by an Argon laser, passively mode-locked by a DODCI jet. In such a cavity, two pulses are propagating in opposite direction, crossing the mod-locking jet at the same time: hence the name of Colliding Pulse Mod-locked (CPM) oscillator.
In order to achieve the shortest possible pulse in such a cavity it is necessary to compensate for the Group Velocity Dispersion (GVD).

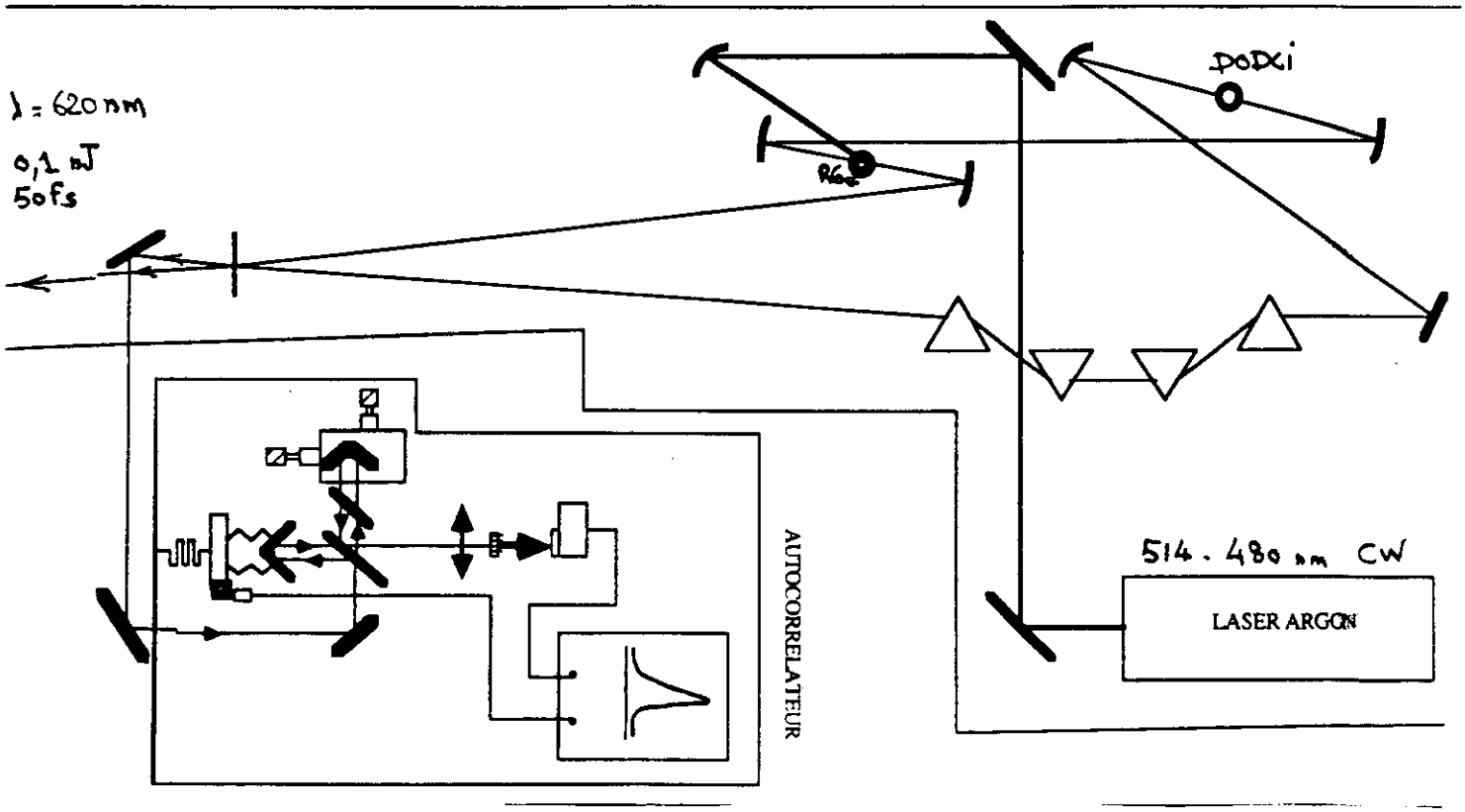


fig. 4

GVD arises from the frequency dependence of the refractive index. The first order GVD term leads to a symmetric temporal broadening. A typical value for the broadening rate is 500 fs/m.nm in Silica at 532 nm. GVD is easily dealt with by using a four-prisms device (fig. 4). It can be shown that for a double prism pair the second derivative of phase is given by:

$$\frac{d^2\phi}{d\omega^2} = \frac{\lambda^3}{2\pi^2 c^2}\left\{4\left[\frac{d^2n}{d\lambda^2} + \left(2n - n^{-3}\right)\left(\frac{dn}{d\lambda}\right)^2\right] l_p \sin\beta \right.$$

$$\left. - 8\left(\frac{dn}{d\lambda}\right)^2 l_p \cos\beta \right\} \qquad (13)$$

see fig. 5 for a definition of l_p and β. For 60° prisms, at minimum deviation, $n = 1.457$, $\omega = 3.1$ rad/fs (615 nm) (13) reduces to

$$\frac{d^2\phi}{d\omega^2}\ (fs^2) = 648 - 32\, l_p \qquad (14)$$

with l_p in cm.
With intra-cavity GVD compensation the output pulse is about 40 fs long, with a repetition rate equal to the round-trip time along the ring (150 MHz) and an energy per pulse of 0.1 nJ typically at 615 nm.

IV.2 Amplifiers

In order to get the intensity necessary to observe multi-photon processes the energy per pulse has to be boosted to the levels of several tens of microjoules. This is obtained by letting the initial pulse go through a number of

amplifying cells pumped by either a frequency doubled q-switched YAG (repetition rate 20 Hz) or a Copper-Vapor laser (10 kHz).
Usually it takes four to five amplifiers to reach an energy level of 1 mJ. Because of the GVD the pulse width at the output of the four stages is about 400 fs. Again this dispersion is compensated by four prisms (or, in a more recent version a two-prism, double pass device). The pulse at 615 nm can be used as is or to generate a supercontinuum.

V. Self-Phase Modulation.

Self-phase modulation (SPM) is the process by which a laser beam propagating in a medium interacts with the medium and imposes a phase modulation on itself. It is the basic process responsible for the supercontinuum generation and for the pulse chirping used in the "Chirped Pulse Amplification" scheme. The aim of this section is to briefly outline the theory of SPM in its most simple form.
The SPM is easily described by the nonlinear wave equation. The physical origin of SPM lies in the nonlinear polarization of the medium at frequency ω induced by the incident electric field $E = \mathcal{E}(z,t)\exp(ikz - i\omega t)$:

$$P_{NL}(\omega) = \chi^{(3)} |E|^2 E \qquad (15)$$

As it is well known, the nonlinear susceptibility $\chi^{(3)}$ is responsible for a large number of physical effects among which is the

the relevant to SPM, namely the non linear index which is usually written as :

$$n(\omega) = n_0(\omega) + n_2 |E|^2 \qquad (16)$$

It can be shown that, in the plane wave approximation, Maxwell equations can be reduced to :

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{v_g} \frac{\partial \mathcal{E}}{\partial t} = i \frac{\omega n_2}{2c} |\mathcal{E}|^2 \mathcal{E} \qquad (17)$$

where $\mathcal{E}(z,t)$ is the complex enveloppe of the electric field E and $v_g = 1/\left(\frac{\partial k}{\partial \omega}\right)$ is the group velocity. Letting $\mathcal{E} = e \exp(i\varepsilon)$, equ (17) reduces to :

$$\frac{\partial e}{\partial z} + \frac{1}{v_g} \frac{\partial e}{\partial t} = 0 \qquad (18)$$

and

$$\frac{\partial \varepsilon}{\partial z} + \frac{1}{v_g} \frac{\partial \varepsilon}{\partial t} = \frac{\omega_0 n_2}{2c} e^2 \qquad (19)$$

The analytical solutions of (18) and (19) are :

$$e(\tau) = e_0 F(\tau)$$

and

$$\varepsilon(z,\tau) = \frac{\omega_0 n_2}{2c} \int_0^z e^2 \, dz' = \frac{\omega_0 n_2}{2c} e_0^2 F^2(\tau) z$$

where e_0 is the amplitude, $F(\tau)$ the pulse enveloppe and τ the local time $\tau = t - z/v_g$.

The electric field, solution of (18) and (19) writes:

$$\mathcal{E}(z,t) = e(\tau) \exp\left[i \frac{\omega_0 n_2}{2c} e_0^2 F^2(\tau) z \right] \qquad (20)$$

The index change, which is time-dependent, induces a time-dependent phase (See fig 5)

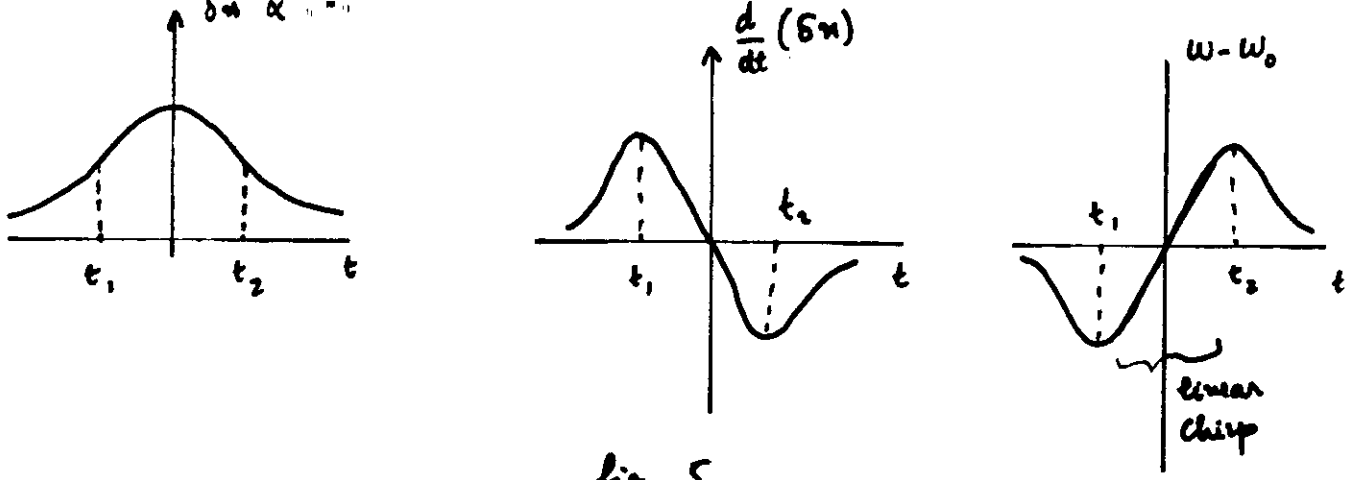


fig. 5

The instantaneous frequency $\omega(\tau)$ is given by :

$$\omega(\tau) = \omega_0 + \delta\omega(\tau)$$

where

$$\delta\omega(\tau) = -\frac{\partial \varepsilon}{\partial \tau} = -\frac{\omega_0}{2c} n_2 e_0^2 z \frac{\partial F^2(\tau)}{\partial \tau}$$

$\delta\omega(\tau)$ is the frequency shift : it is proportional to the derivation of the pulse enveloppe. The spectrum of SPM pulses is obtained by taking the Fourier transform of the complex enveloppe. For a gaussian pulse $F(\tau) = \exp[-\tau^2/2\tau_0^2]$ it can be shown that the maximum frequency extent is

$$\Delta\omega_{max} \simeq \frac{\omega_0 n_2 e_0^2 z}{c \tau_0} \qquad (21)$$

$\Delta\omega_{max}$ is inversely proportional to the pulse duration and proportional to the non linear index n_2.

The chirp (temporal distribution of frequency during the pulse) is characterized by a coefficient defined by the phase relation

$$\varepsilon = C \tau^2 \qquad (22)$$

For a gaussian pulse and linear approximation

$$F(\tau) = \exp[-\tau^2/2\tau_0^2] \simeq 1 - \frac{\tau^2}{\tau_0^2} \qquad (23)$$

c becomes :

$$c = \left(\frac{\omega_0}{2c}\right)\left(\frac{n_2 \mathcal{E}_0^2 z}{\tau_0^2}\right) \qquad (24)$$

VI. Supercontinuum generation

Supercontinuum generation takes place in gases, solids and liquids. Non linear effects in solids are very effective. However, damage thresholds are rather low and for ultrashort, high-intensity effects, liquids are more suitable. One of the most favourite liquid medium used to generate supercontinuum is H_2O or D_2O. With incident 100 fs pulses at 615 nm a cylindrical cell, 2-cm long generate continuum extending below 310 nm on the anti-Stokes side and to the near IR region on the Stokes side. For the purpose of multiphoton experiments (see lecture II) a narrow band is selected out of this continuum by an interference filter. The energy in this band is very low (0.1 nJ regime) and must be cranked up to several microjouls to be suitable for multiphoton transitions. This is achieved by two amplifier Dye cells, the second one being used in double pass. It is necessary, to obtain a high intensity, to be able to tightly focus the resulting beam. This makes necessary to spatially filter the beam between the two amplifier stage using a lens pin-hole combination. The output level is thus raised to 10 microjoules.

VII. Other Schemes for short, intense pulses generation

The set up which has just been described (CPM + dye amplifications) is suitable for mJ amplification. This is due to the high emission cross-section ($\sigma_e \sim 10^{-16}$ cm^2) and the low energy storage capability of the dyes (mJ/cm^3). The same holds for excimer amplifiers. If one wants to reach very high peak power, solid-state amplification is the most suitable one. In this section, I briefly describe the multiterawatt system built at Saclay and the KrF* laser developped at the University of Illinois at Chicago.

VII.1 Multiterawatt, picosecond Nd-Glass laser system

Solid state amplification has proved to be very efficient because of the high storage capacity (joule/cm^3) and saturation intensity (J/cm^2) available in Nd-glass or YAG or the new materials like Ti: Sapphire or Alexandrite. In order to take advantage of this high energy storage capability, it is necessary to keep the pulses long enough to avoid damage caused by too much intensity and wave-front distorsion due to non linear effects in the amplifying rods. On the other hand, very high intensity demands short to ultrashort pulses. The solution to this problem is the so-called Chirped Pulse Amplification (CPA). This technique was first developped by G. Mourou. A schematic diagram of the experimental arrangement is shown on fig. 6.

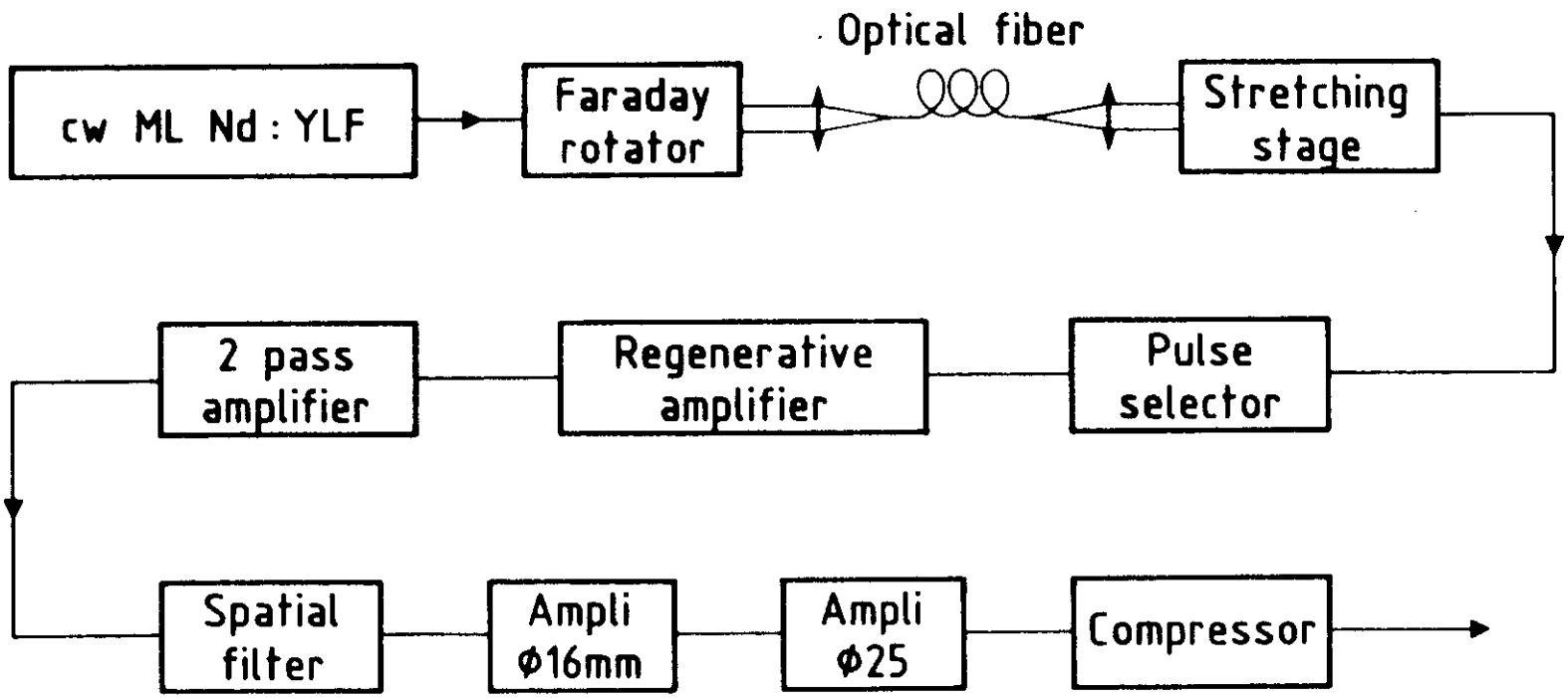

Optical fiber
cw ML Nd : YLF
Faraday rotator
Stretching stage
2 pass amplifier
Regenerative amplifier
Pulse selector
Spatial filter
Ampli ϕ16mm
Ampli ϕ25
Compressor

Fig. 6

The output pulse train from a cw-mode-locked Nd-YLF oscillator (1053 nm, 50 ps, 0.2 Å, 100 MHz) is coupled via a microscope objective to a single mode optical fiber. As the pulses propagate along the 800 m of the fiber, they undergo SPM and are broadened to 250 ps and chirped over 35 Å. At this point, the pulses are stretched to 1 ns in a 4-pass stretching stage using two antiparallel gratings associated with a telescope. (1740 lines/mm). It can be shown that such a device has a positive GVD, as the fiber, and contributes to a stretching of the pulse too. A single stretched pulse is selected out from the train and amplified to 5 mJ in a regenerative amplifier (7 mm Nd-phospate glass, 0.1 Hz). The multipass amplification narrows the spectral bandwidth to 14 Å and, consequently, the pulse duration narrows to 0.5 ns. This effect has the advantage of selecting those frequencies with a linear chirp (see fig 5) suitable for the linear compression. The pulse is then amplified to 30 mJ and spatially filtered. The final stage consists of a 16 mm- and a 25 mm-diameter rods used single-pass to reach 2.2 Joules. Before entering the compression stage, the beam diameter is enlarged to 33 mm to keep intensity below the gratings damage threshold. The autocorrelation measurement yields a 0.6 ps duration at the output of the compression stage, corresponding to a 2.5 TW peak power. (1 shot/minute). After focussing this laser produces an intensity in excess of 10^{17} W cm^{-2}.

VII.2 The KrF laser

Excimer amplifiers combine a broad gain-bandwidth product and a high threshold for the onset of non linear processes. Furthermore in a number of experiments in plasma physics investigations, the short wavelength (248 nm) is well suited, due to a dramatic frequency dependence (ω^{-4}). Recently, the Chicago group has reported measurements performed on a large-aperture KrF* system capable of output power over 400 GW. The laser is based on the following scheme: a 248 nm seed pulse is generated from a subpicosecond dye laser at 745 nm frequency doubled and mixed (745 nm → 372 nm → 248 nm). The seed pulses have an energy of about 1 μJ. A first amplifier stage (KrF*) has a gain of about 500. After spatial filtering, the seed pulse has an energy of 150-200 μJ. The beam is then collimated and expanded to a 10 cm-diameter and injected into the final stage amplifier ("Prometheus"). This system has a 2.5 m-long discharge and the gaz, pre-ionized by an X-ray gun, is flowed at a rate sufficient to clear the discharge volume about 5 times between pulses. The gain is about 2.9% cm^{-1}. An output energy of 250 mJ is reached with a seed pulse energy of 200 μJ. The pulse duration was measured by the two-photon fluorescence technique to be 600 fs. The beam minimum section, after focusing by an aspheric, 60 mm-focal length mirror, has a diameter of 1.7 μm. Therefore, across this

focus the average intensity is $2.10^{\sim} W cm^{-2}$. To our knowledge this is the highest intensity achieved by such "small-scale" systems. For instance, a Neon atom submitted to such an intensity loses 8 electrons. Charge spectra of produced ions clearly show Ne^{8+} while only Ne^{4+} was observable at $10^{16} W cm^{-2}$.

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