Laser-induced plasma generation and characterizations. "Basic Concepts"

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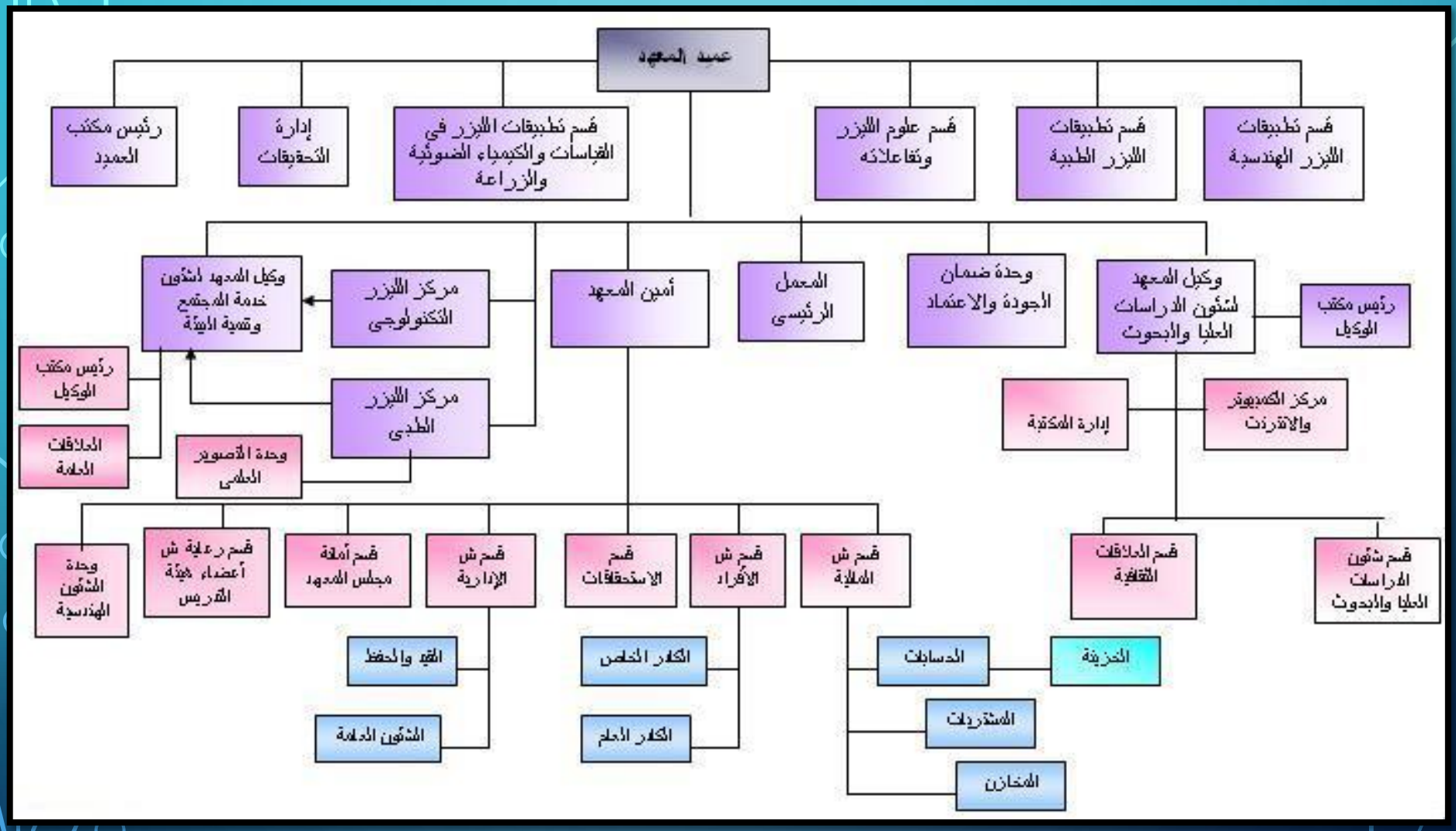
Outline of the of part I

1. Lasers and their properties
2. Mechanisms involved in laser material interaction
3. Plasma formation and its evolution (temporal , spatial)- Other effects (laser shielding, shock wave formation)
4. Ultrafast lasers

Cairo University founded on 1908

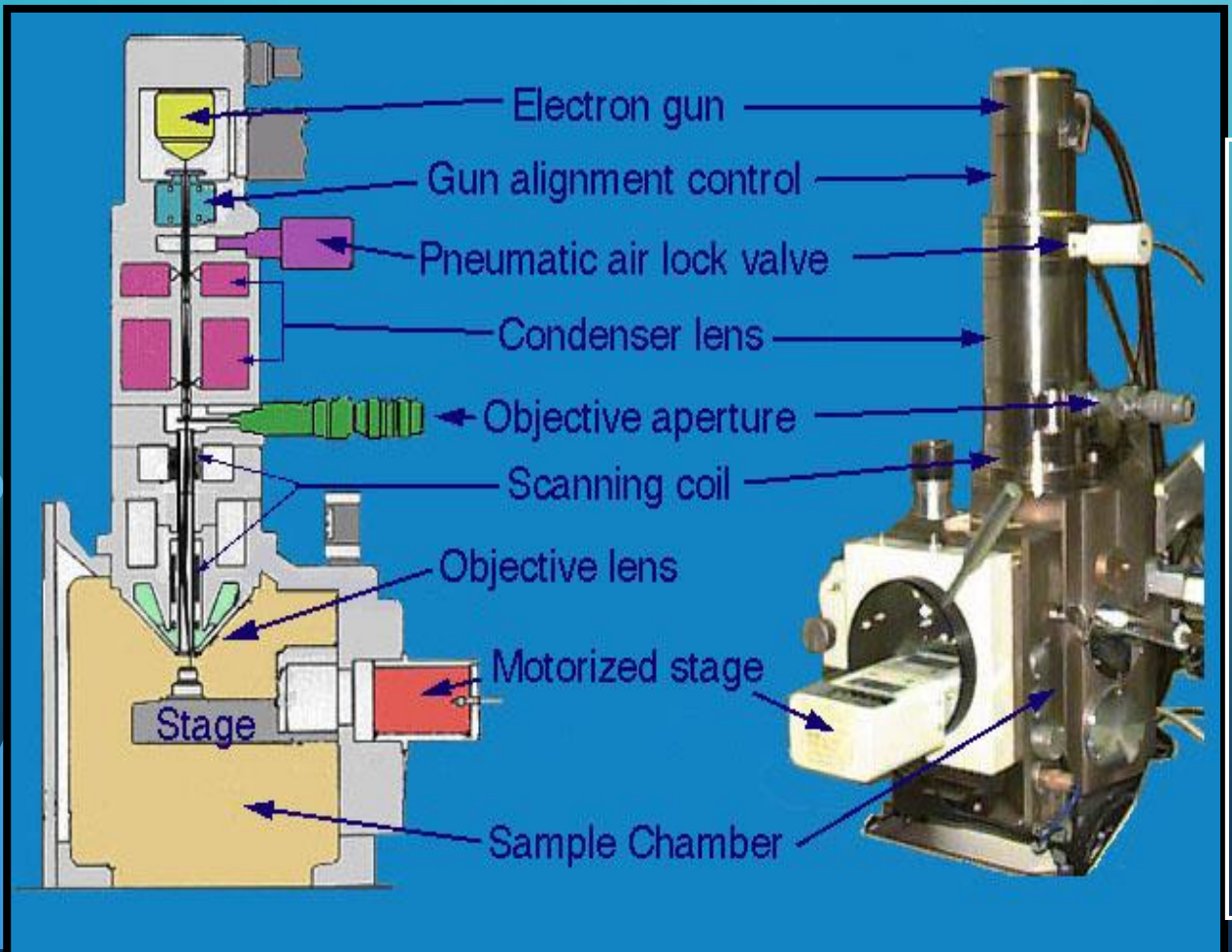
NATIONAL INSTITUTE OF LASER NILES WAS ESTABLISHED IN 1994.

الهيكل الأداري للمعهد



بعض الاجهزة بالمعمل

الميكروسكوب الاليكتروني
Scanning electron Microscope

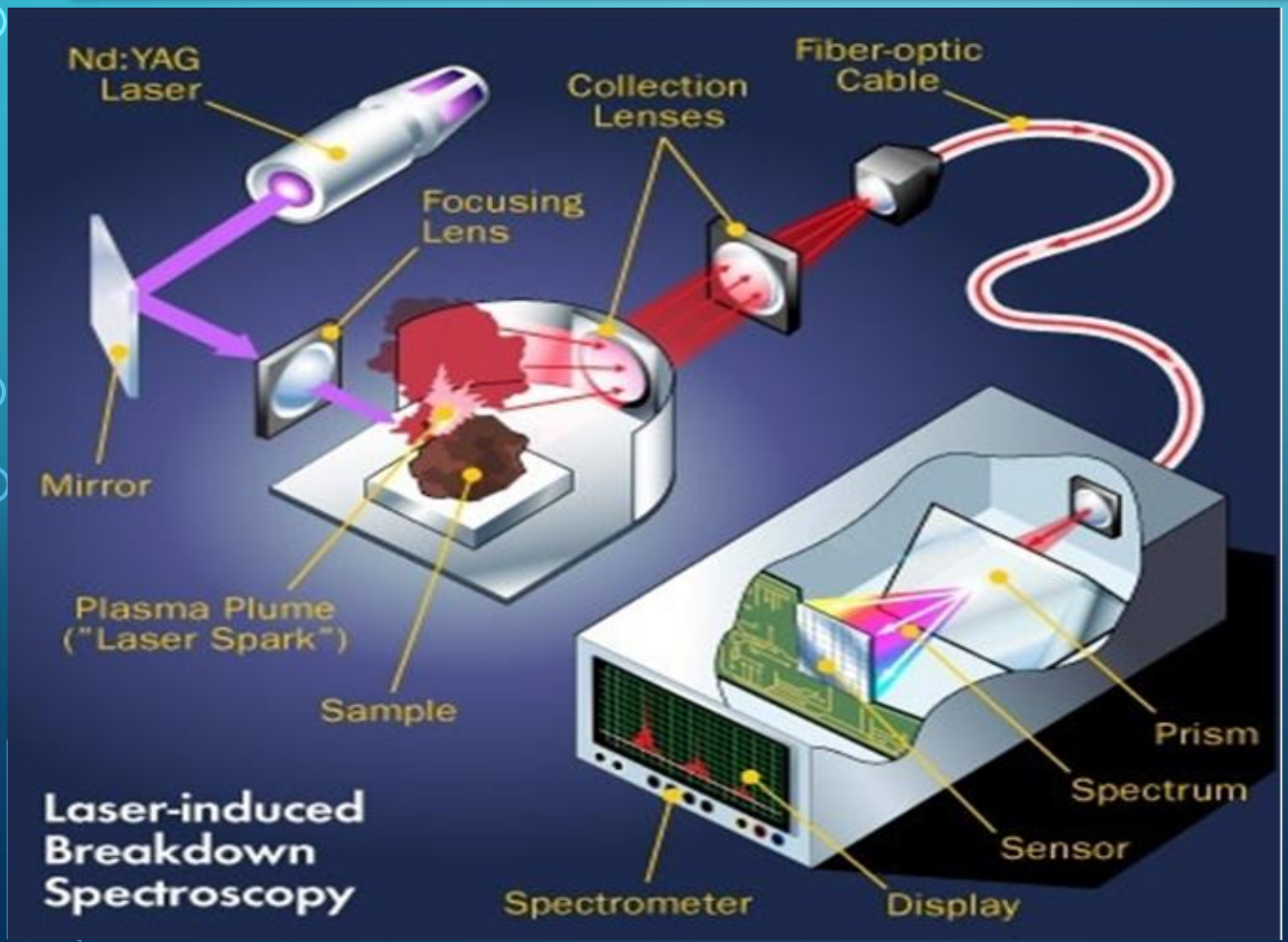


- electron gun (filament)
- electromagnetic optics
- scan coils
- sample stage
- detectors
- vacuum system
- computer hardware and software (not trivial!!)

بعض الاجهزة بالمعمل

Laser Induced Breakdown Spectroscopy

LIBS: **L**aser **I**nduced **B**reakdown **S**pectroscopy system



LIBS is an analytical method by which one can determine (qualitatively and quantitatively) the elemental composition of solid, liquid or gas samples.

بعض الاجهزة بالمعمل

High energy Nd: YAG laser source

A 30 Hz Nd:YAG laser (Continuum Model NY81) 5ns, 500 mJ 1064 nm, and KDP for SHG, THG are available

بعض الاجهزة بالمعمل

Moderate energy Nd: YAG laser source

A 10 Hz Nd:YAG laser (Continuum Model Surelite I) 5ns, ~ 300 mJ 1064 nm, and KDP for SHG, THG and FHG are available.

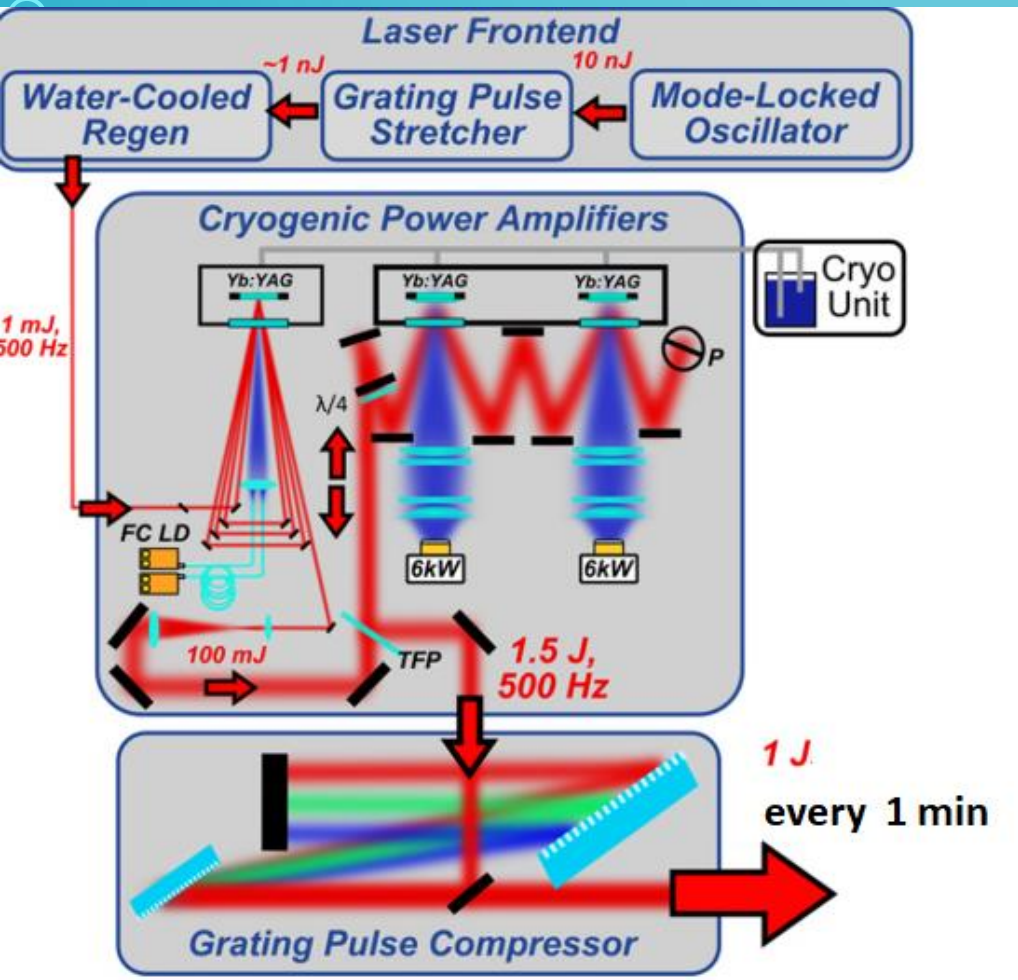
بعض الاجهزة بالمعمل

Complete PLD system (pulsed laser deposition)

Complete PLD system (pulsed laser deposition) includes; pulsed Nd:YAG lasers , turbo high vacuum pump, rotary pump, a vacuum chamber with optical windows for diagnostic purposes and proper optics.

بعض الاجهزة بالمعمل

"ND:Glass" picosecond laser 1-2 joules per pulse



بعض الاجهزة بالمعمل

Femtosecond laser oscillator

Ti:sapphire laser oscillator pumped by Argon ion laser – with output of 100 femtosecond at 80 MHz and energy of 10 nJ

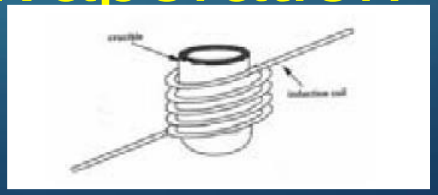
بعض الاجهزة بالمعمل

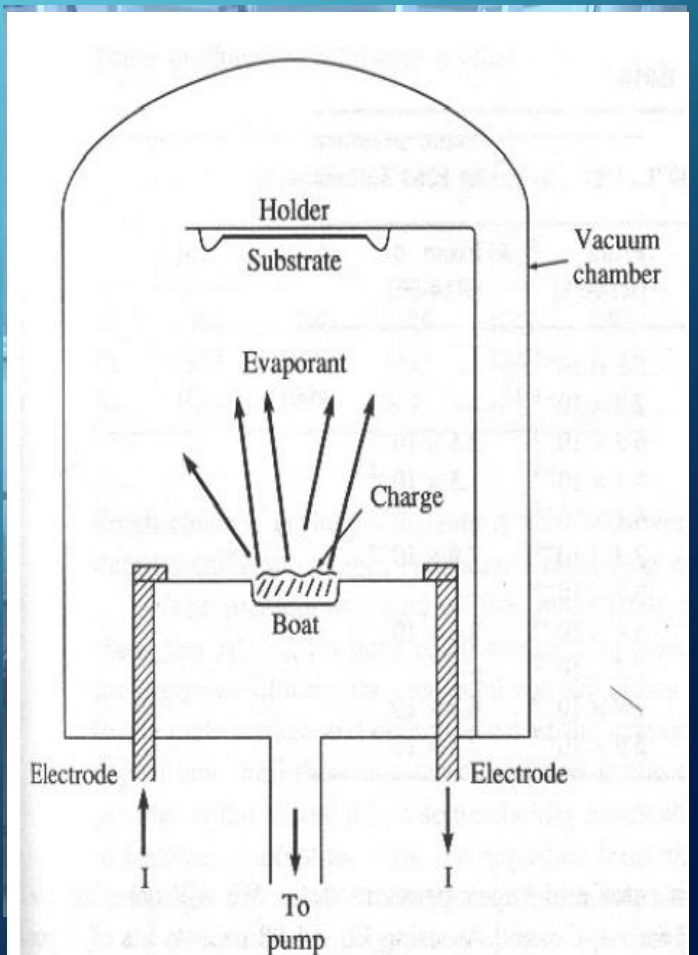
Thin Film Deposition Coating Unit

Thermal Induced Evaporation

Resistance Evaporation sputtering

Induction Heated Thermal Evaporation

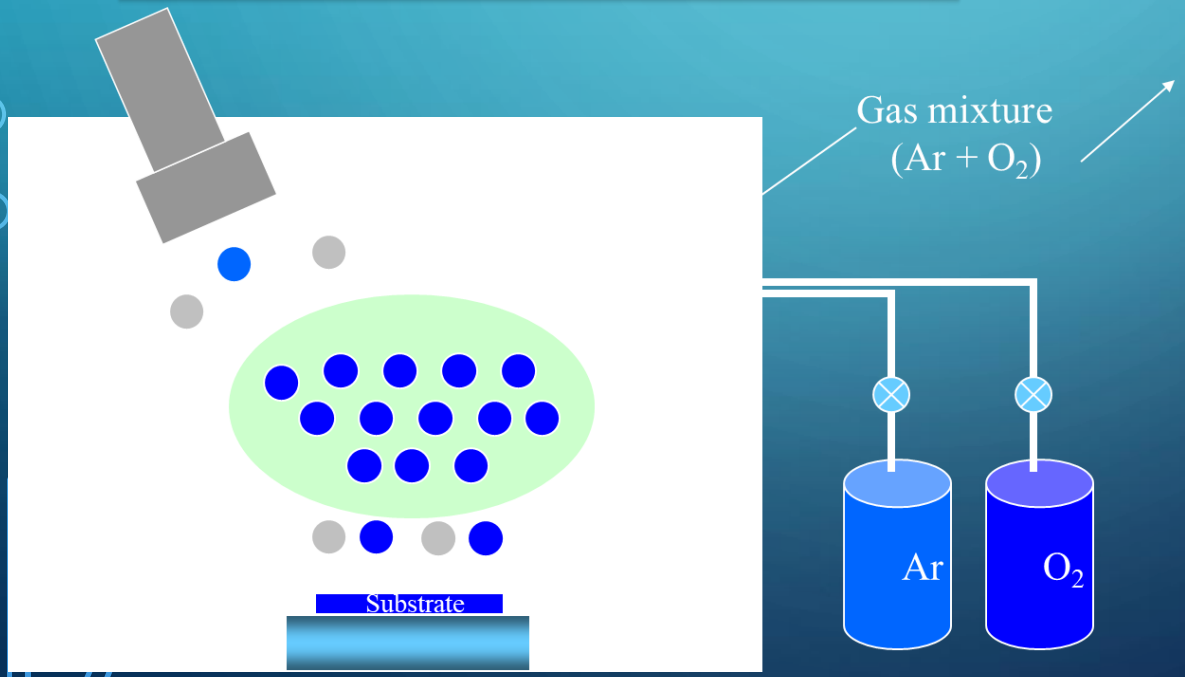




بعض الاجهزة بالمعمل

Thin Film Deposition
Coating Unit

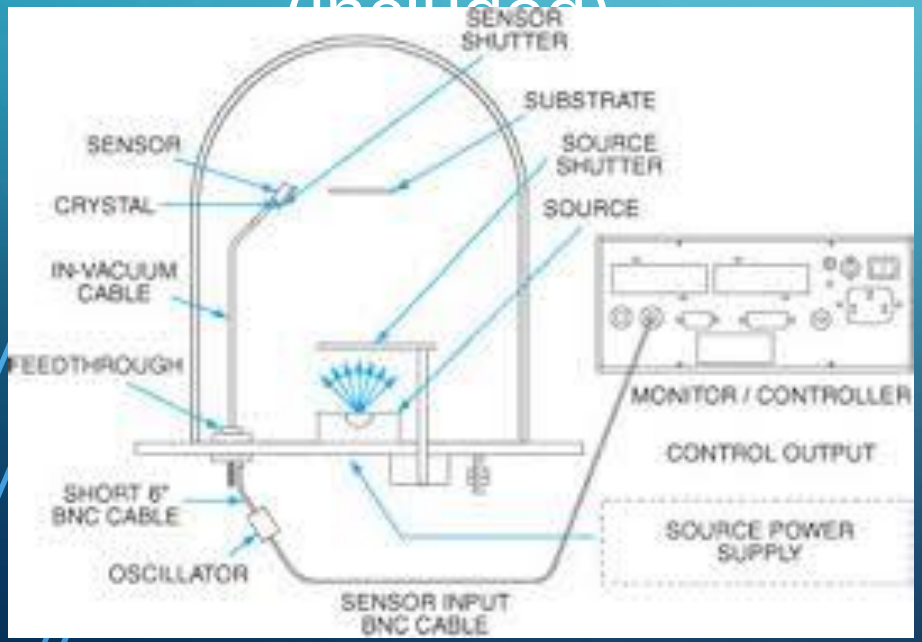
Electric Sputtering
الترسيب بالرش الكهربي



بعض الاجهزة بالمعمل

Thin Film Deposition Coating Unit

thin film thickness measuring unit (included)



The very useful gravimetric technique to measure both deposition rate and film thickness involves quartz crystal oscillator. It is based on using the thickness shear mode of piezoelectric quartz crystal. Films as thin as 10 nm up to ~ 100 microns can be measured.

What is a Laser?

- Light Amplification by Stimulated Emission of Radiation.
- Laser properties:

1. High Intensity and Brightness: many orders of magnitude higher than a traditional light source.
2. High monocromaticity (temporal coherence): Typically with laser stabilized He—Ne laser stabilized Sodium Lamp

Typical Values for Brightness

Source	Power	Beam divergence	Area	Brightness
Hg arc	10 kW	4π sr	1 cm^2	$\sim 1000\ W\ cm^{-2}\ sr^{-1}$
Sun	4×10^{26} W	4π sr	$2.5 \times 10^{23}\ cm^2$	$\sim 130\ W\ cm^{-2}\ sr^{-1}$
He–Ne laser	10 mW	3×10^{-4} rad	0.1 cm^2	$\sim 10^6\ W\ cm^{-2}\ sr^{-1}$
Ruby laser	10 MW	5×10^{-3} rad	1 cm^2	$\sim 4 \times 10^{11}\ W\ cm^{-2}\ sr^{-1}$
Specially designed high brightness Nd–glass laser	4 GW	4×10^{-5} rad	10 cm^2	$\sim 2 \times 10^{17}\ W\ cm^{-2}\ sr^{-1}$

3. High Spatial Coherence and High Directionality : very important to achieve a little spot in focusing the beam on a target, so to obtain an high irradiance

Lasers

Light **A**mplification by **S**timulated **E**mission of **R**adiation

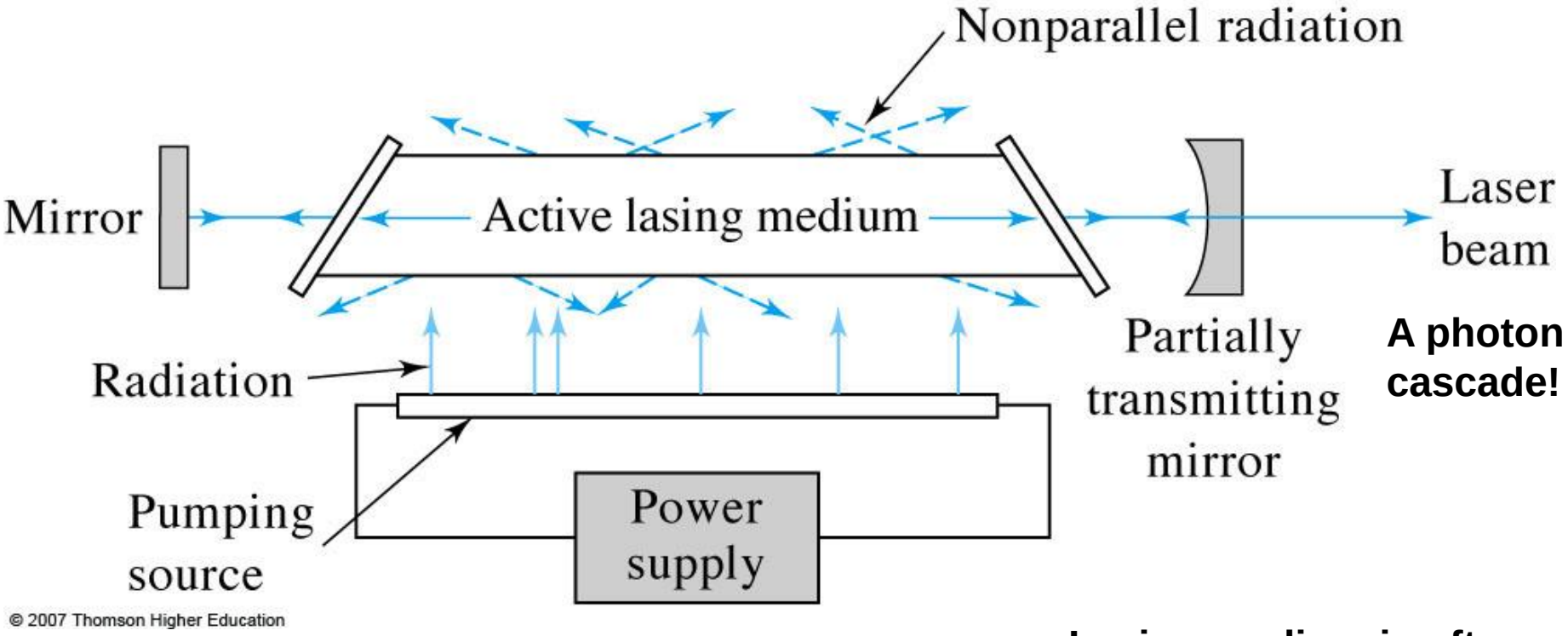
- Intense light source
- Narrow bandwidth (small range $\lambda < 0.01$ nm)
- Coherent light (in phase)

Lasers

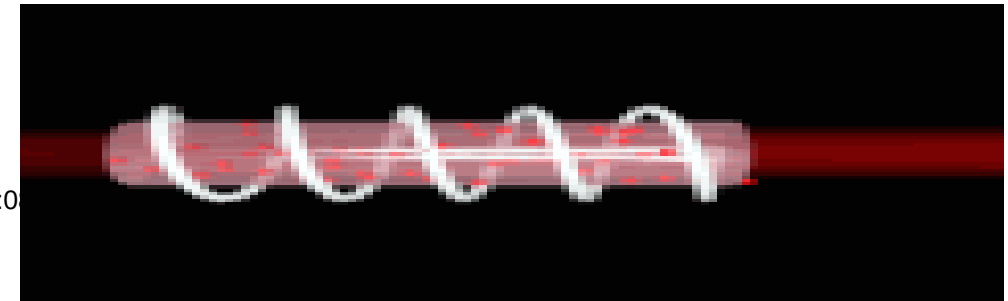
Light **A**mplification by **S**timulated **E**mission of **R**adiation

- Pumping
- Spontaneous Emission
- Stimulated Emission
- Population Inversion

Laser design



A photon cascade!

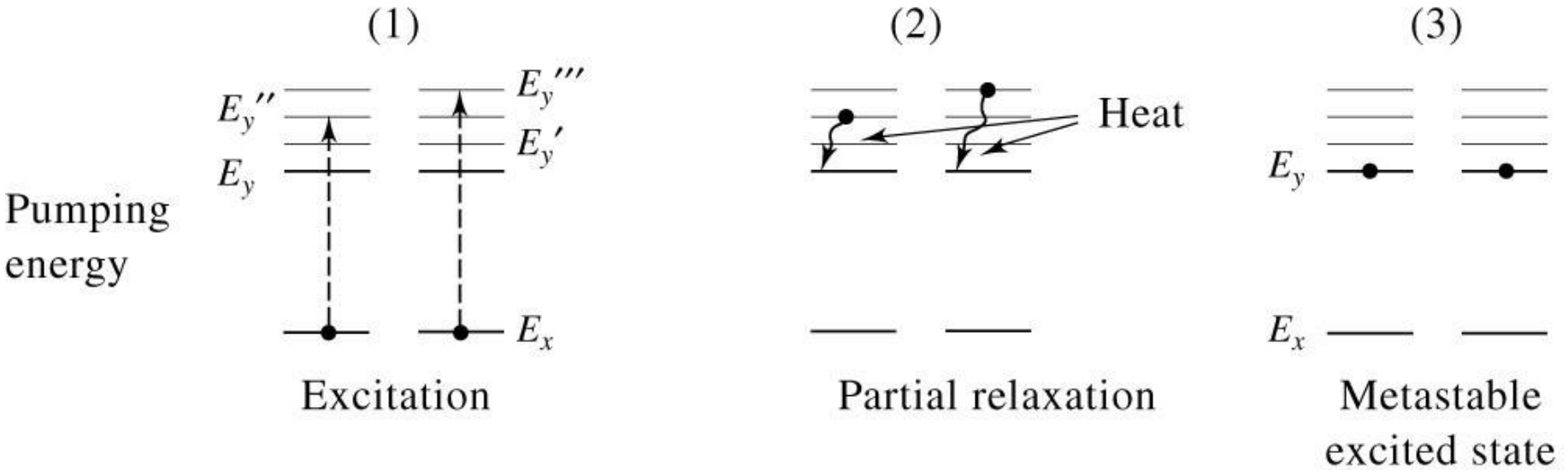


Lasing medium is often:

- **a crystal, like ruby**
- **a dye solution**
- **a gas or plasma**

Pumping

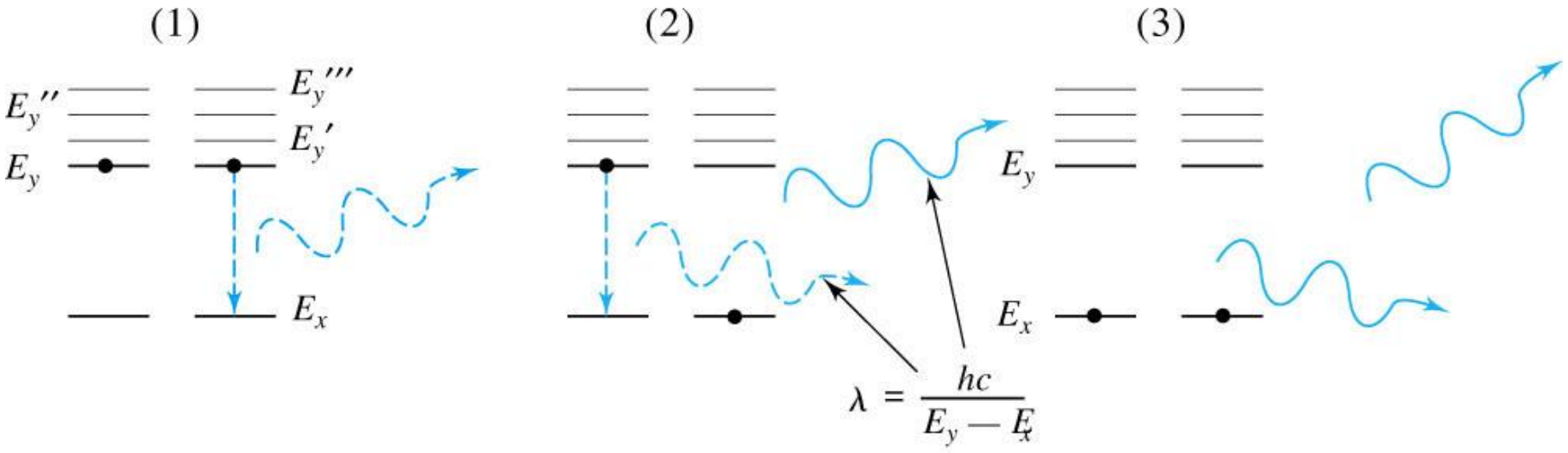
- Generation of excited electronic states by thermal, optical, or chemical means.



(a) Pumping (excitation by electrical, radiant, or chemical energy)

Spontaneous emission or relaxation

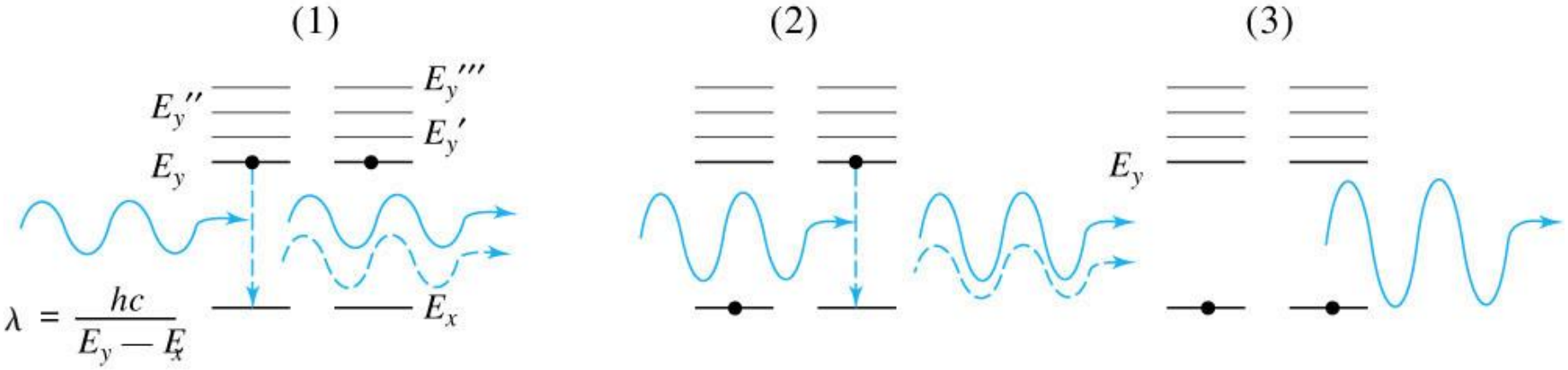
- Random in time
- No directionality
- Monochromatic (same λ), but incoherent (not in phase)
- Solid vs. dashed line – 2 different photons



(b) Spontaneous emission

Stimulated emission

- The excited state is struck by photons of precisely the same energy causing immediate relaxation
- Emission is **COHERENT**
  - Emitted photons travel in same direction
  - Emitted photons are precisely in phase



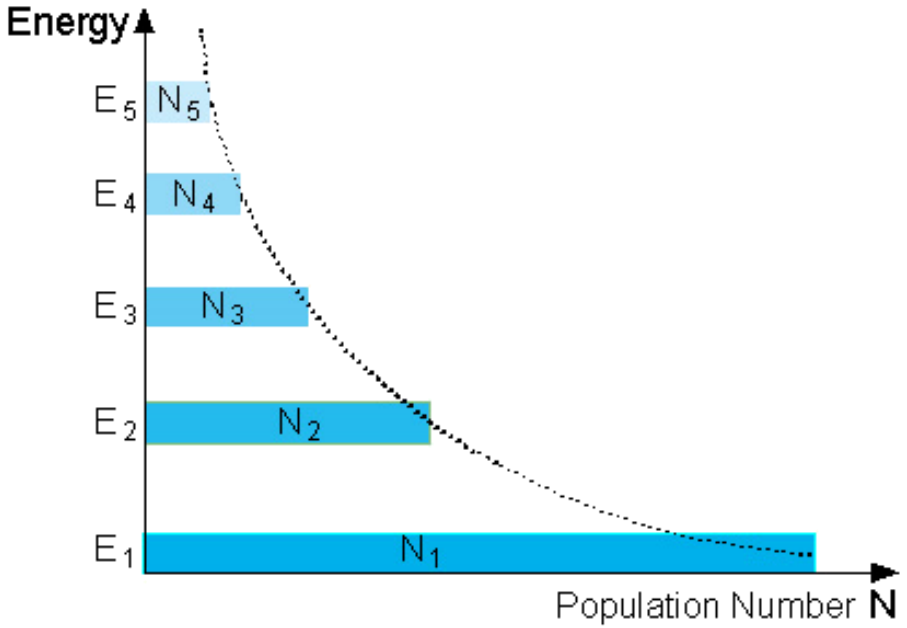
(c) Stimulated emission

Population at thermal equilibrium

- **The relationship between atom population at energy levels at equilibrium is described by Maxwell-Boltzman Law.**

$$N_1 = \frac{g_1}{g_0} N_0 \; e^{-\frac{h\upsilon}{KT}}$$

Where T is the temperature in Kelvin and there is a thermal equilibrium at T, g is the statistical weight which represent the different ways of distribution of atoms all have the same energy (degeneracy).



Example:

- Calculate the ratio of the population numbers (N1, N2) for the two energy levels E2 and E1 when the material is at room temperature (300^{o}K), and the difference between the energy levels is 0.5 [eV]. What is the wavelength (λ) of a photon which will be emitted in the transition from E2 to E1?

- **Solution:**

When substituting the numbers in the equation, we get:

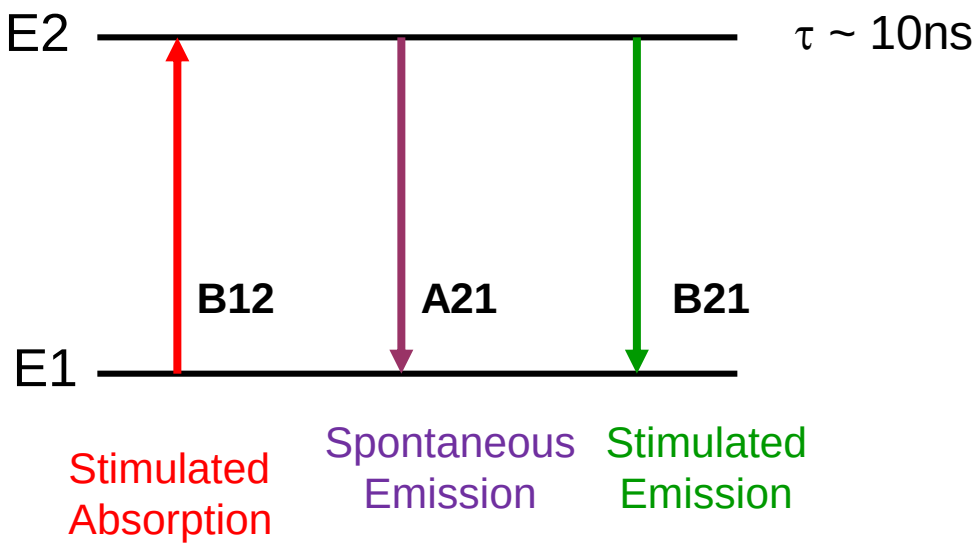
- $$\frac{N_2}{N_1} = \exp\left(-\frac{E_2 - E_1}{k_B \cdot T}\right) = \exp\left[-\frac{(0.5 \cdot eV) \cdot \left(1.6 \cdot 10^{-19} \cdot \frac{J}{eV}\right)}{\left(1.38 \cdot 10^{-23} \cdot \frac{J}{K}\right) \cdot (300K)}\right] = 4 \times 10^{-9}$$

- This means that **at room temperature, for every 1,000,000,000 atoms at the ground level (E1), there are 4 atoms in the excited state (E2) !!!**

- To calculate the **wavelength:**

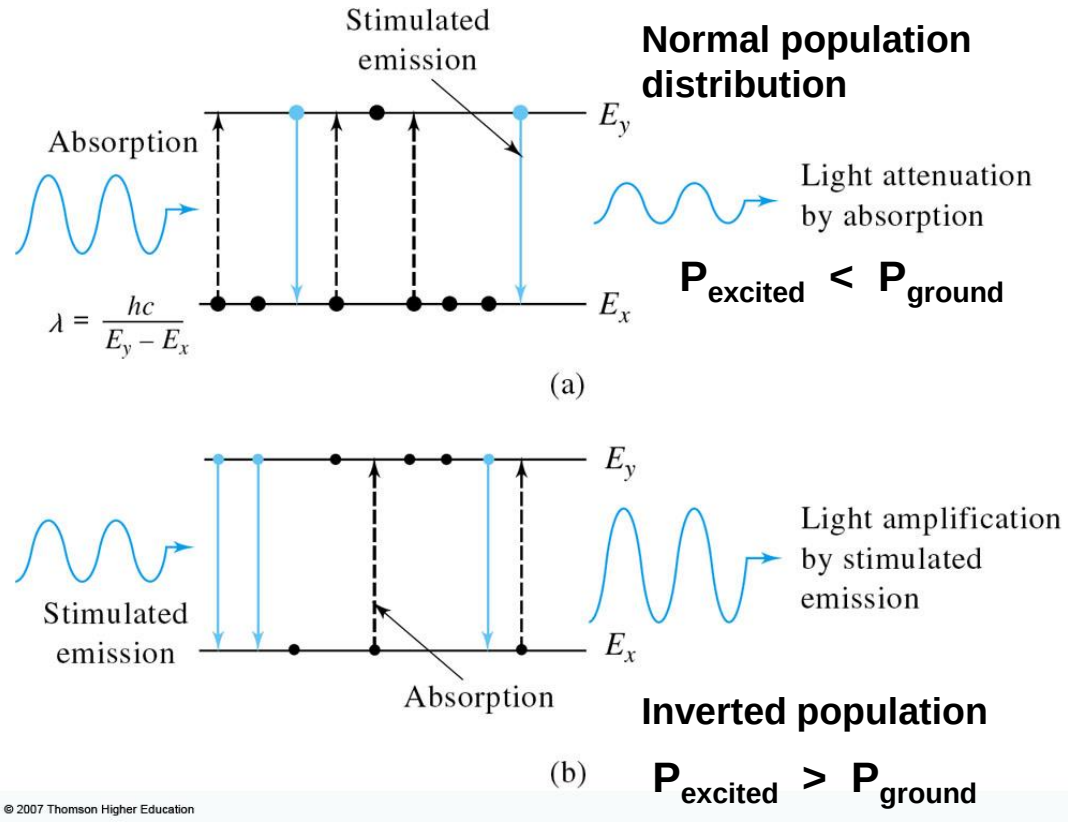
$$\lambda = \frac{h \cdot c}{\Delta E} = \frac{(6.626 \cdot 10^{-34} \cdot J \cdot sec) \cdot \left(3.10^{8} \cdot \frac{m}{sec}\right)}{(0.5 \cdot eV) \cdot \left(1.6 \cdot 10^{-19} \cdot \frac{J}{eV}\right)} = 2.48 \cdot \mu m$$

- **This wavelength is in the Near Infra-Red (NIR) spectrum.**



Population inversion

- When the population of excited state species is greater than ground state, an incoming photon will lead to *more* stimulated emission *instead* of absorption.



Normal population distribution

$P_{excited} < P_{ground}$

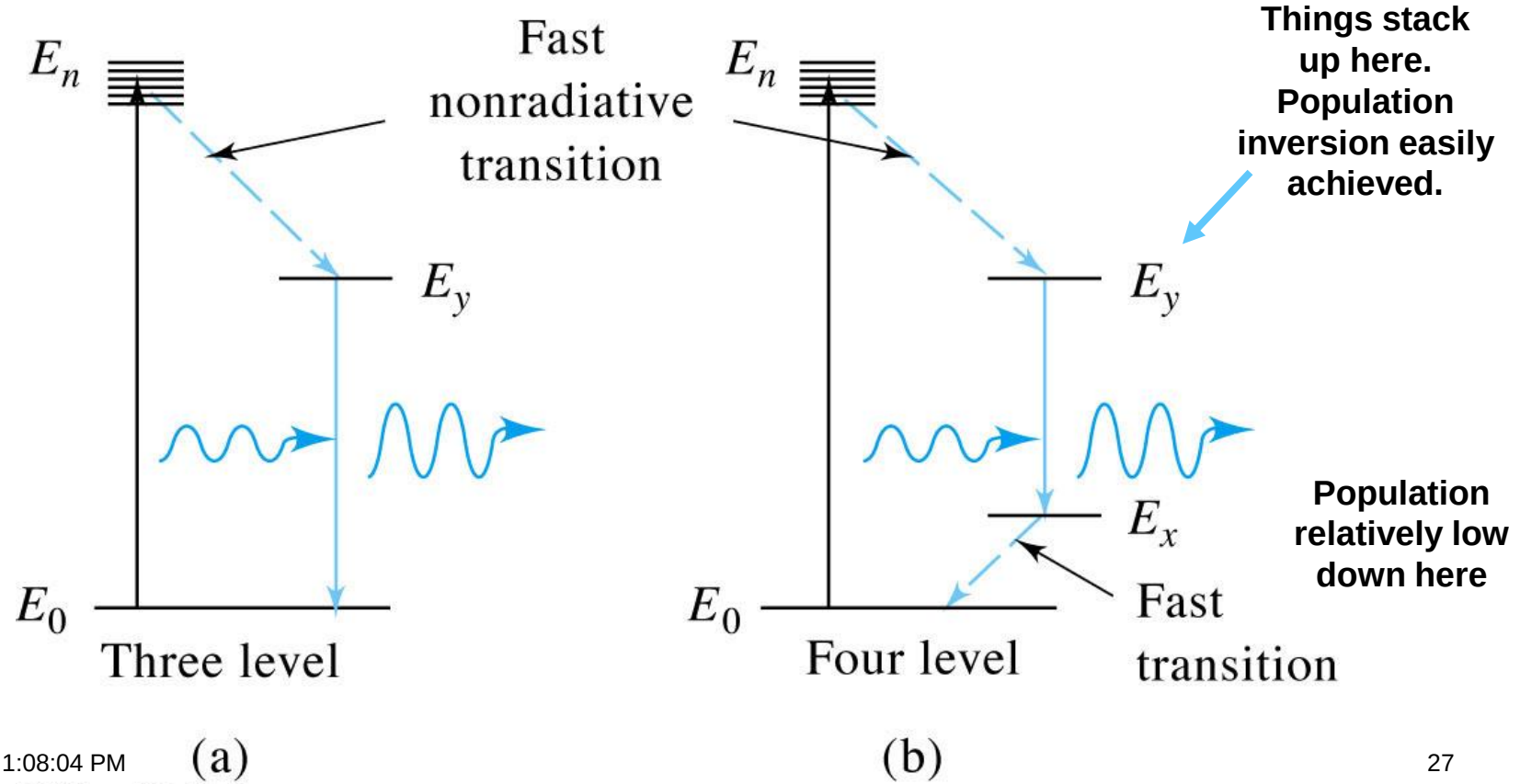
Inverted population

$P_{excited} > P_{ground}$

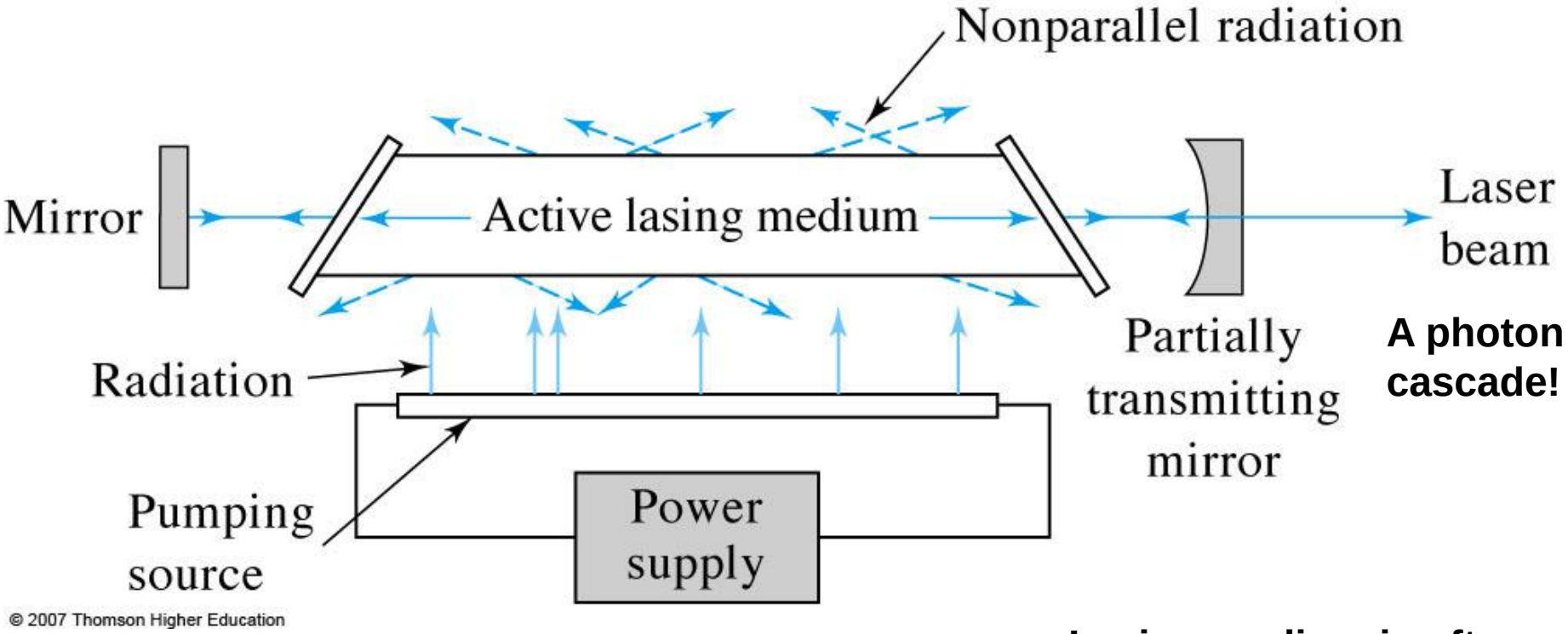
Laser action

3- and 4-state lasers

- Population inversion easier in 4-state system



Laser design



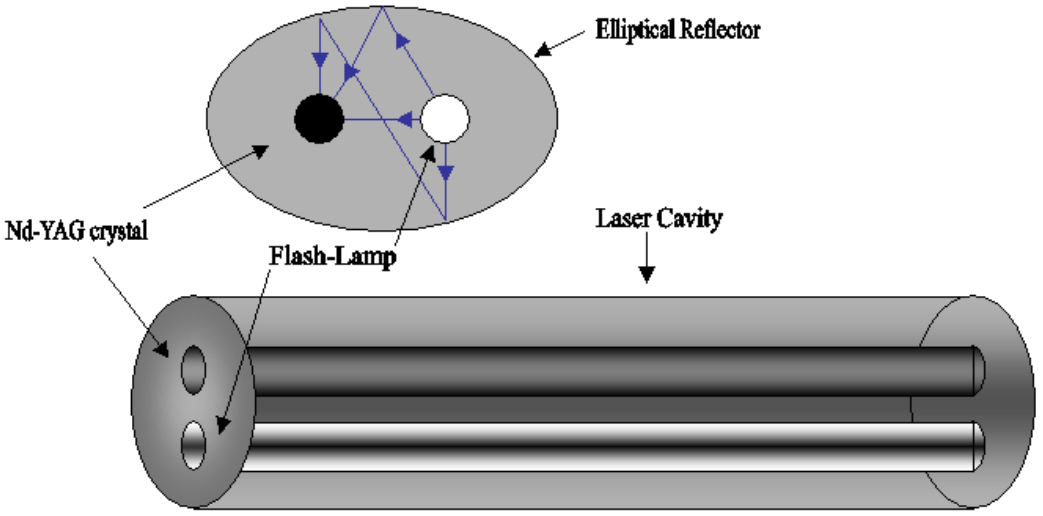
A photon cascade!

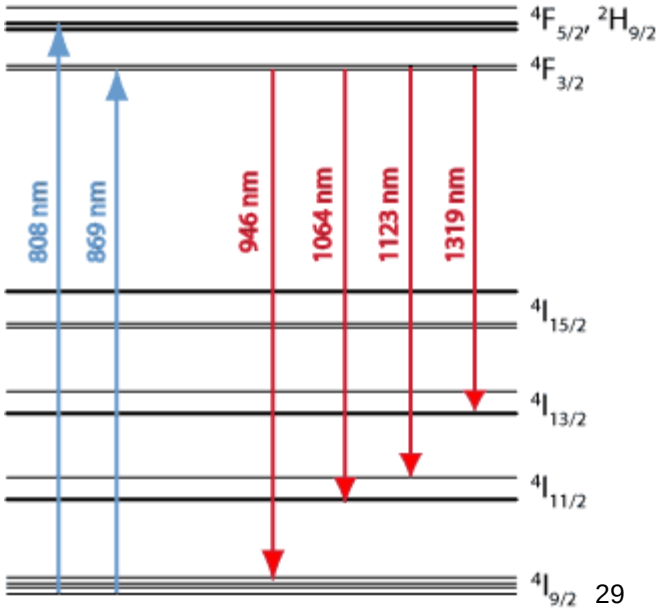
Lasing medium is often:

- **a crystal, like ruby**
- **a dye solution**
- **a gas or plasma**

Continuous wave laser sources

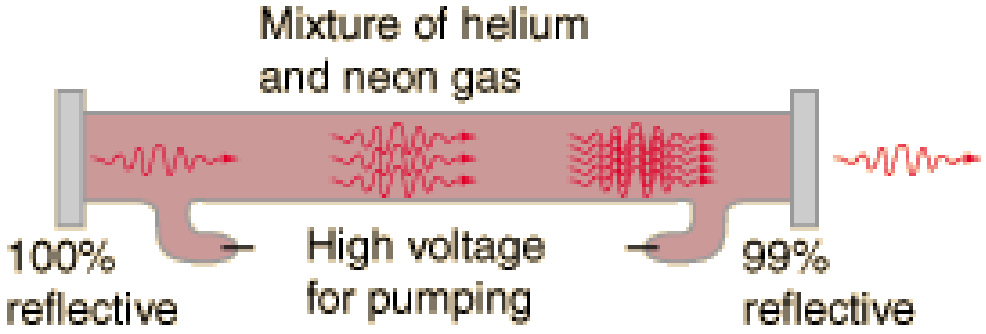
- Nd^{3+}:Yttrium aluminum garnet (YAG: $Y_3Al_5O_{12}$)
  - Solid state
  - **1064 nm**, **532 nm**, 355 nm, 266 nm
- The GTE Sylvania Model 605, uses a Nd-YAG laser rod set in a "double elliptical“ reflector, is pumped by two 500-W incandescent lamps, and is limited to a low order mode by an aperture in the laser cavity.

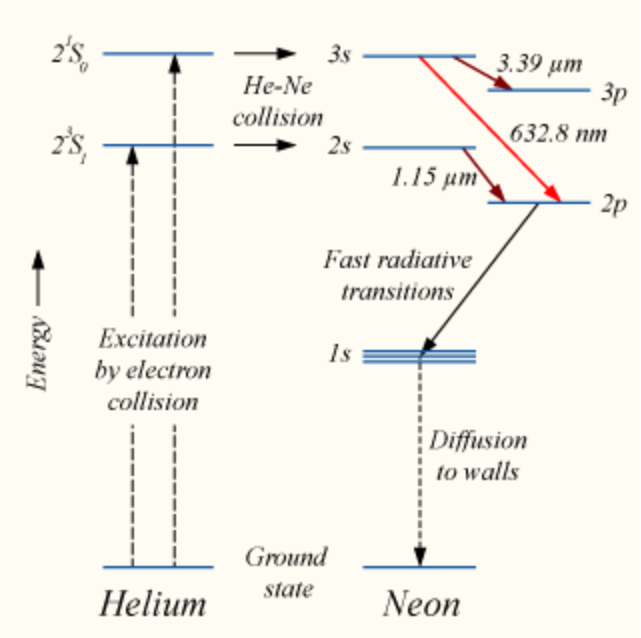




Continuous wave laser sources

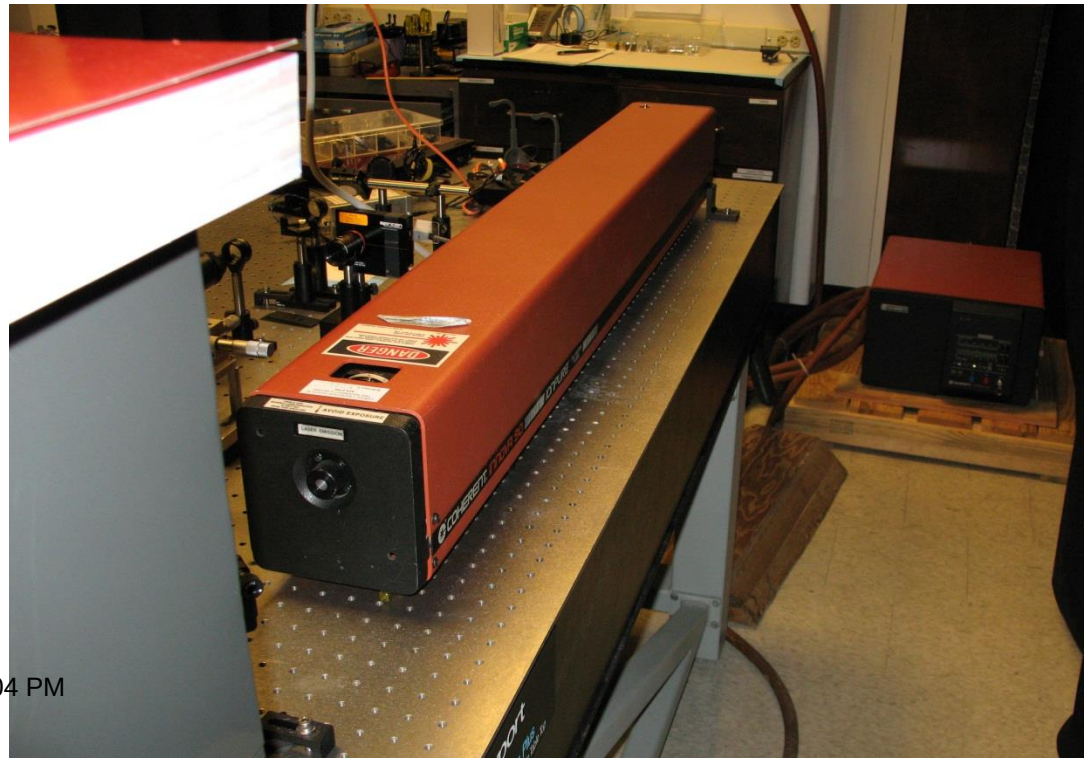
He-Ne laser is a 10:1 mixture, at low pressure in a glass tube. The gas mixture is mostly helium, so that helium atoms can be excited and further collide with neon atoms, exciting them so it may radiate 632.8 nm. Without helium, the neon atoms would be excited mostly to lower excited states and radiate non-laser lines.





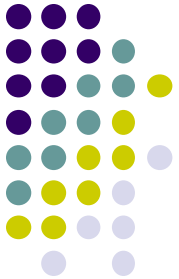
Continuous wave laser sources

- Ar+
  - Gas laser, but emission comes from ions
    - Uses lots of electrical power to generate ions
  - 351.1 nm, 363.8 nm, 454.6 nm, 457.9 nm, 465.8 nm, 476.5 nm, **488.0 nm**, 496.5 nm, 501.7 nm, **514.5 nm**, 528.7 nm, 1092.3 n



Coherent Innova 90
Up to 5 W of output!
~100x my laser pointer

Other continuous wave laser sources

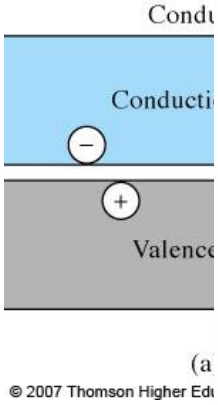


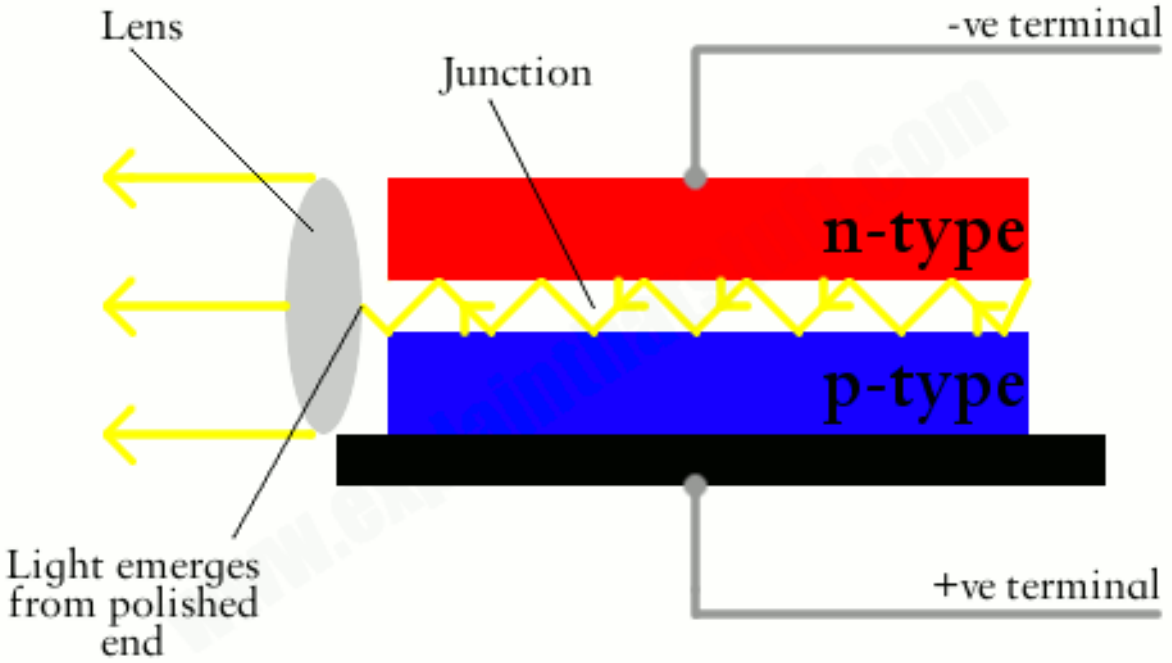
- Cu vapor
  - 520 nm
- HeCd
  - **440 nm**, 325 nm
- Dye lasers

Pulsed lasers sources

- Nd:YAG
  - Solid state
  - Often nanosecond pulses
  - **1064 nm, 532 nm**, 355 nm
- Ti:sapphire
  - Solid state—often pumped by Nd:YAG
  - Tunable output aroudn 800-1200 nm
  - Produces femtosecond pulses
- Nitrogen
  - Gas
  - **337 nm**
- Excimer lasers (gas mixtures; excited state is stable)
- Tunable dye lasers (λ is selective within limits)

Laser diodes



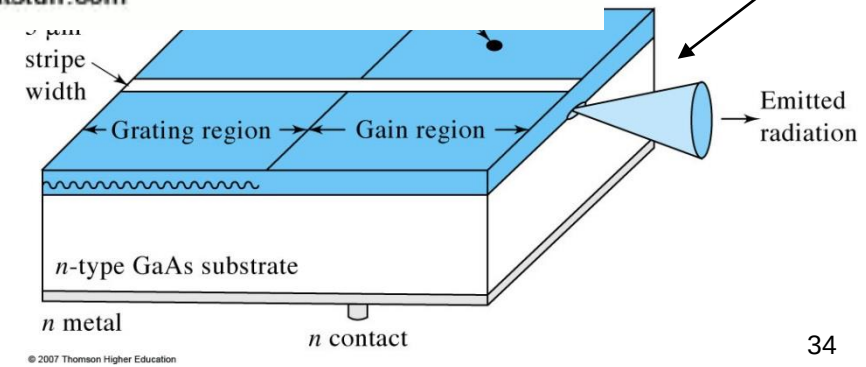


Band gap energy, E_g

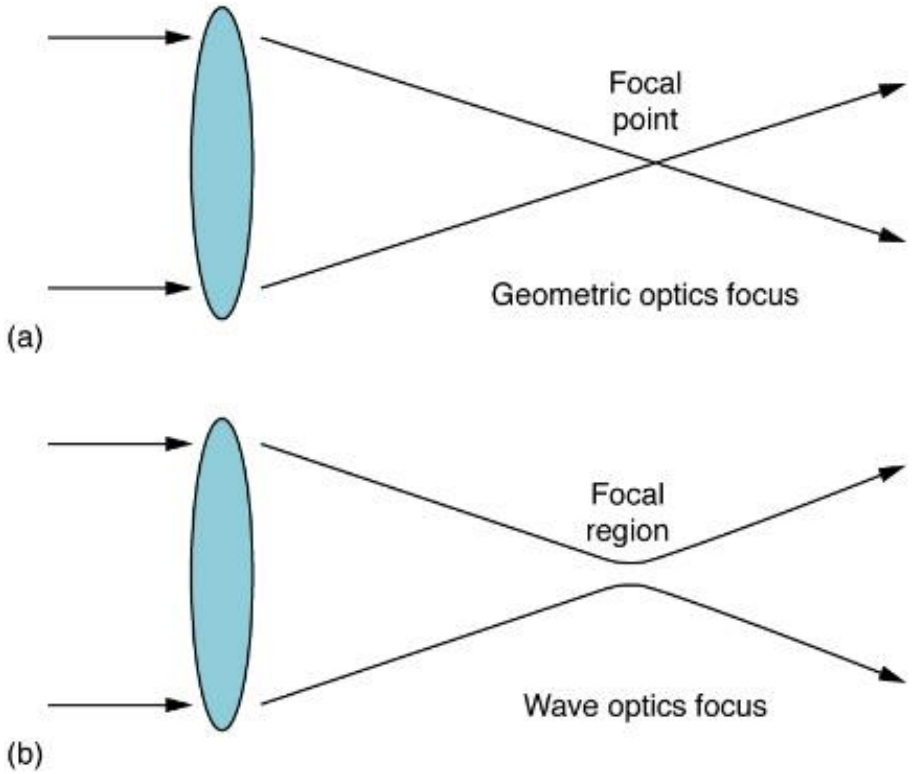
www.explainthatstuff.com

- Used in CD and DVD players (not very strong)
- Wavelengths now available from IR to near UV regions

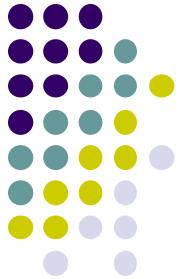
Resonant Cavity emits At 975 nm



Focusing Laser Beam



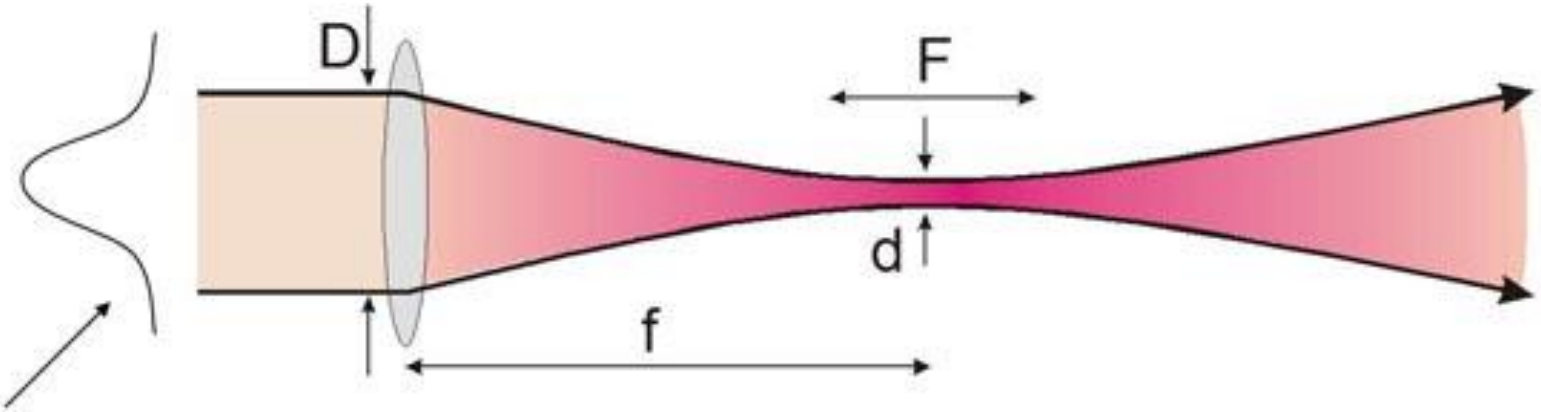
Focusing Laser Beam



Laser beams are typically very well collimated. Diffraction causes light waves to spread transversely as they propagate, and it is therefore impossible to have a perfectly collimated beam. The diffraction-limited divergence angle of a Gaussian beam with diameter D and wavelength λ is $\Theta = 4 \cdot \lambda/\Pi \cdot D$. As an example, for an Argon laser emitting a 1 mm wide beam at 515 nm wavelength, the divergence is about 0.66 mrad, ie, the beam spreads by 1.3 mm over a distance of 1 meter.

Using a lens or a concave mirror with focal length f, a laser beam can be focused to a spot with a diameter $d = (4 \cdot f/\Pi \cdot D)\lambda$. The depth of the focal region is: $F = (8 \cdot f^2/\Pi \cdot D^2)\lambda$. With a f=25 mm lens the same Argon laser beam can be focused to a spot of 16 μm in diameter, having a focal depth of 820μm. It must be emphasized though that exact definition of the spot size depends on the beam profile, which varies in various configurations of laser cavities.

Focusing Laser Beam



$I(r) = I_0 \exp(-2r^2/w^2) = I_0 \exp(-8r^2/D^2)$ Numerical aperture NA = sin (D/2f)

For a Gaussian laser beam:

Diameter of the focal spot

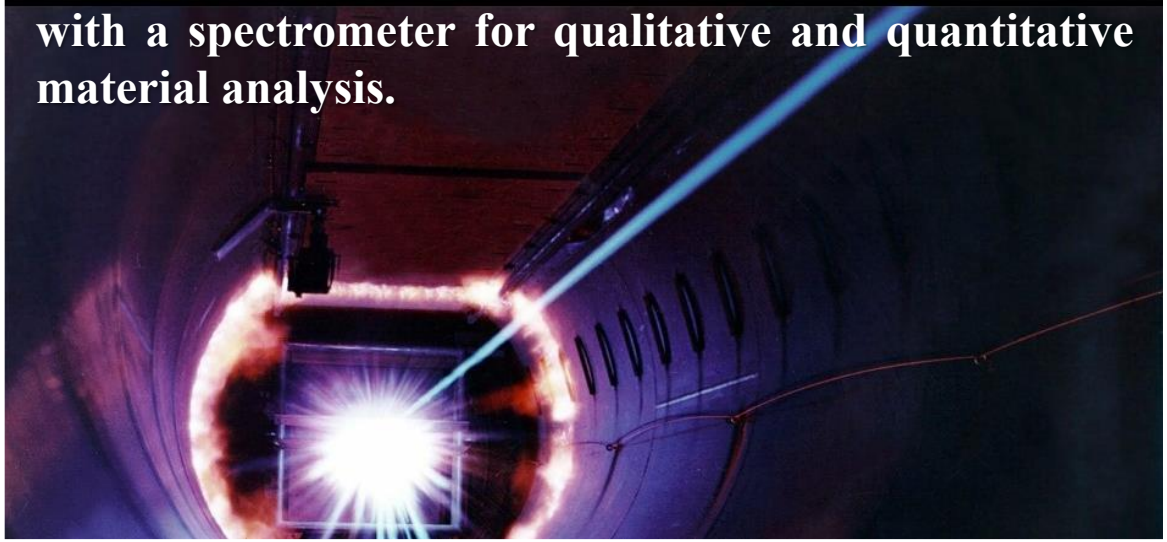
$$d = \frac{4 \cdot f}{\pi \cdot D} \lambda \cong \frac{2 \cdot \lambda}{\pi \cdot NA}$$

Focal depth

$$F = \frac{8 \cdot f^2}{\pi \cdot D^2} \lambda \cong \frac{2 \cdot \lambda}{\pi \cdot NA^2}$$

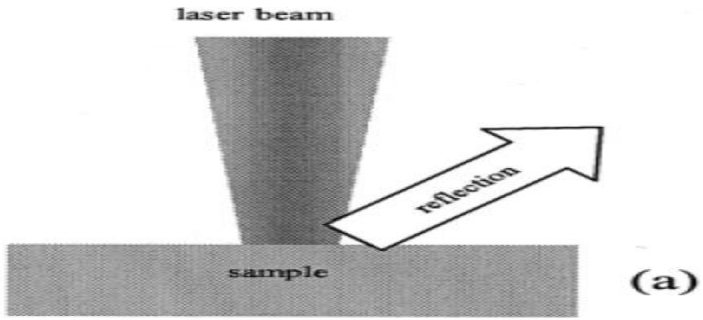
Laser Induced Plasma Spectroscopy

Laser Induced plasma Spectroscopy (LIPS) is a type of atomic emission spectroscopy that uses a laser to vaporize or ablate a microscopic layer of a sample's surface. The resultant plasma caused by this laser ablation process emits light as it cools. This is then followed by collecting the light and then analyzing it with a spectrometer for qualitative and quantitative material analysis.

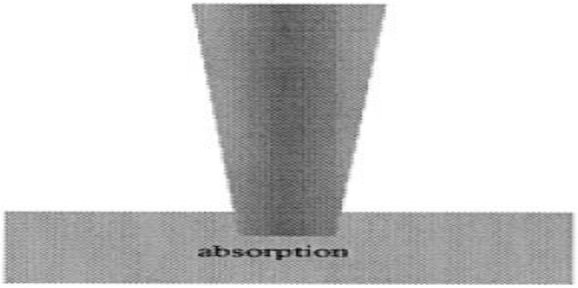


Laser interaction with matter

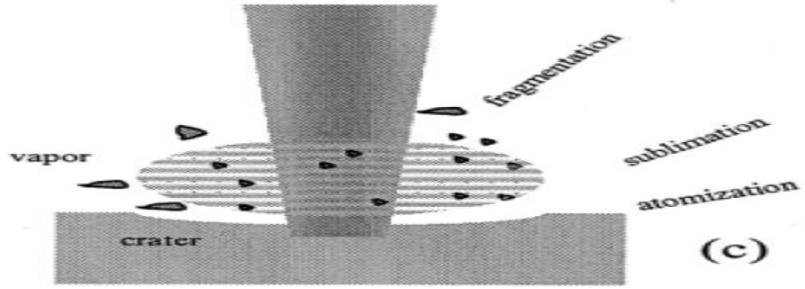
Mechanisms involved in Laser interaction			
	solid	gas	liquid
1- Reflection	●		●
2- Absorption	●	●	●
3- Melting	●		
4- Vaporization	●		●
6- Plasma formation	●	●	●
7- Laser - Plasma Interaction	●	●	●
8- Shock wave formation	●	●	●
9- Hydrodynamic Expansion		●	
10- Ejection of solid target	●		


laser beam
reflection
sample

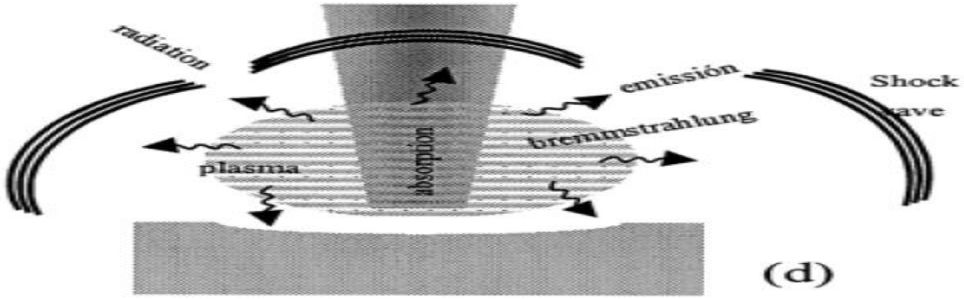
(a)


absorption

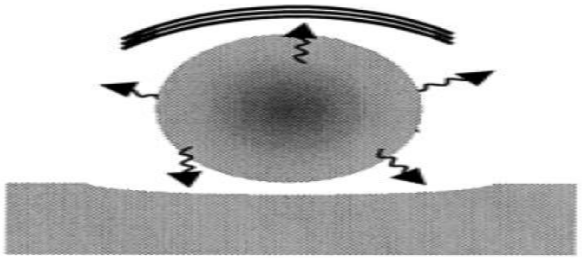
(b)


fragmentation
vapor
sublimation
atomization
crater

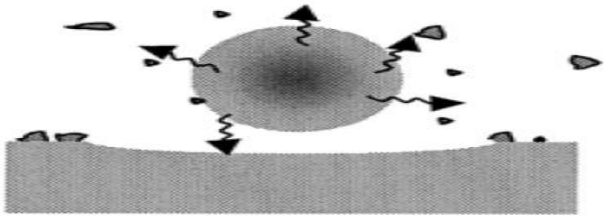
(c)


radiation
emission
Shock
wave
bremsstrahlung
plasma
absorption

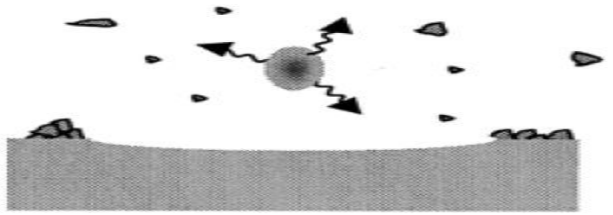
(d)



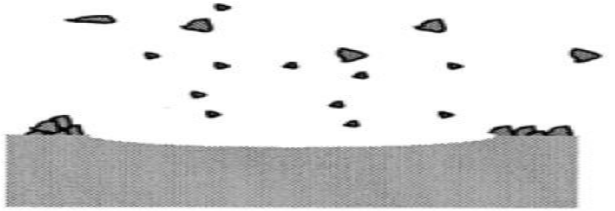
(e)



(f)

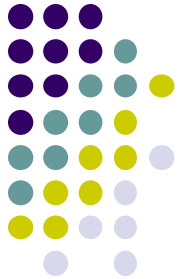


(g)



(h)

Why LIBS?



- Atomic emission spectroscopy
  - Provides basic chemical information of species (Can analyze every element)
- Can allow for qualitative measurement.
- Real-time – no sample preparation
- Simple and performed on solid, liquid, gas, and aerosol species.
- Cheaper than other analytical techniques

Parameters affecting LIBS

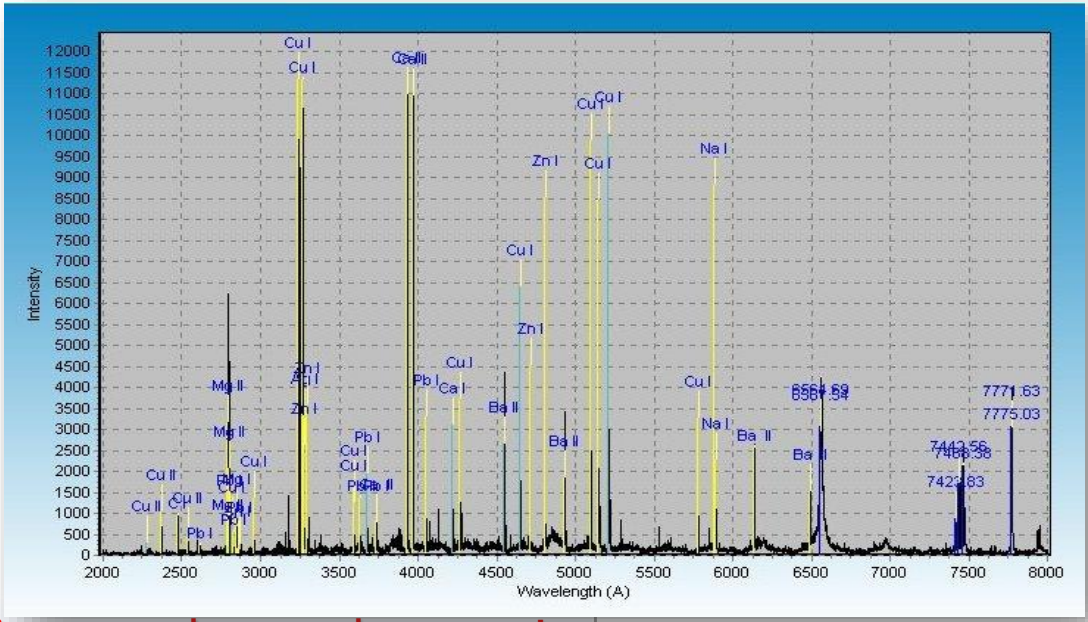
- Laser energy and irradiance
- Pulse width
- Laser wavelenght (UV, IR, …)
- Ambient conditions (composition, pressure)
- Matrix of the sample
- Thermal properties of the sample
- Focusing conditions

A complete model is missing

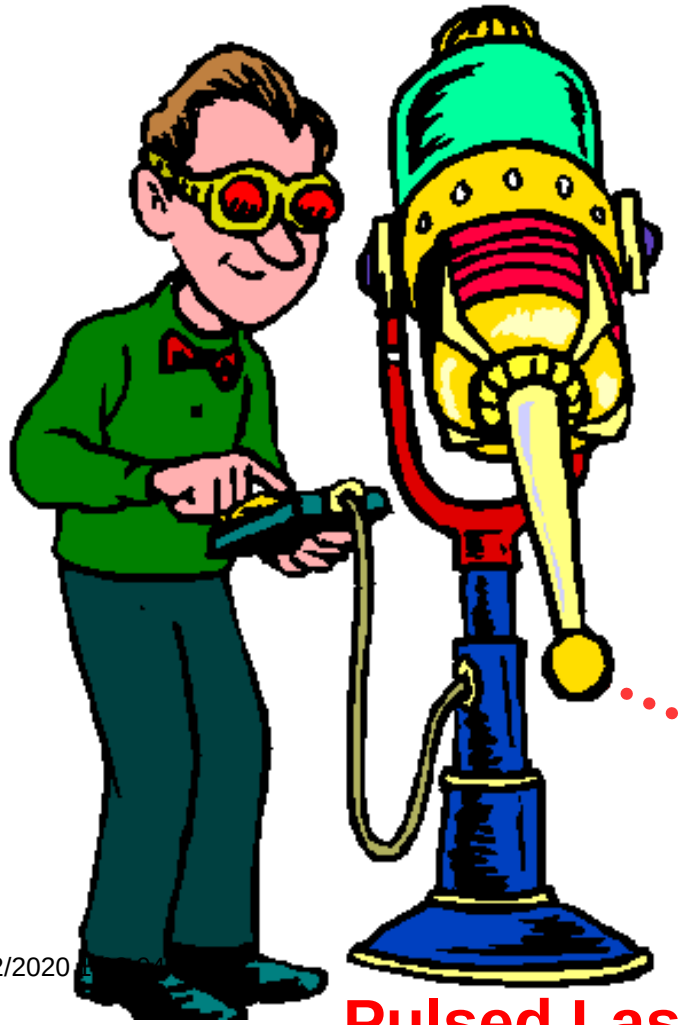
In order to understand the best experimental conditions for target sampling and analytical study,

we must check case by case

LIPSBasic



As a laser beam is focussed on the sample, the local temperature quickly rises until exceed the breakdown condition ,inducing the plasma formation



Plasma Plume

Pulsed Laser

Plasma and its parameters

- Highly ionized cloud of particles containing neutral atoms, Ions, and free electrons
- Can have high electron densities (up to $10^{24}/cm^3$) and temperatures (up to 10^8 K) **(In our case $N_e \approx 10^{18}/cm^3$ and $T_e \approx 10000$ K)**

1- Plasma temperature

For plasma in **local thermodynamic equilibrium (LTE),** the population density of atomic and ionic electronic states is described by a Boltzmann distribution.

$$I = \frac{hc}{4\pi\lambda}.N(T).\frac{A_{ki}g_k}{U(T)}.\exp(-\frac{E_k}{KT})$$

I = the emitted spectral line intensity

N(T) = total density (Ions + neutrals)

λ = the wavelength

k and h are Boltzman and planks constants, respectively

A_{ki} = the transition probability

g_k = the statistical weight for the upper level k

U(T) = partition functions

E_k = the excited level energy

T is the electron temperature in Kelvin (in LTE all temperatures are assumed to be equal, i.e. $T_e \approx T_{ion} \approx T_{plasma}$)

Reformulating Boltzmann Eqn. gives

$$\ln\frac{I\lambda}{A_{ki}g_k} = -\frac{1}{KT}E_k + \ln\frac{C\cdot F}{U(T)}$$

- where *F is* an experimental factor and *C* is the species concentration.
- By plotting the left hand side of this equation vs. the excited level energy $E_{k,}$ the plasma temperature can be obtained from the slope of obtained straight line.

2- Plasma Density

$$N_e \approx \left(\frac{\Delta\lambda_{FWHM}}{2.W}\right).10^{16}$$

N_e is the electron density (in cm^{-3})

1. Stark-broadening results from the
2. interaction of an emitter with an electric field
   of a charged particle at a distance r
   causing a shift in energy that is linear in the
3. field strength.

depends on the electron density N_e.

$\Delta\lambda_{FWHM}$ = is the fundamental width at half maximum of the measured line.

W = the electron impact parameter (stark broadening value).

Boltzmann plot

$$y = m x + q_s$$

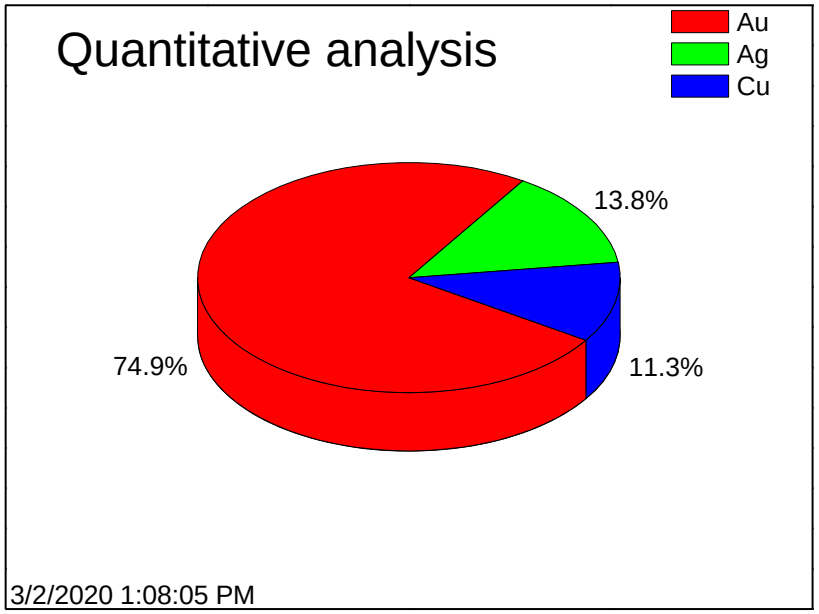
$$\ln\frac{I\lambda}{A_{ki}g_k} = -\frac{1}{KT}.E_k + \ln\frac{C\cdot F}{U(T)}$$

$$y = \ln\frac{\overline{I_\lambda^{ki}}}{g_k A_{ki}}$$

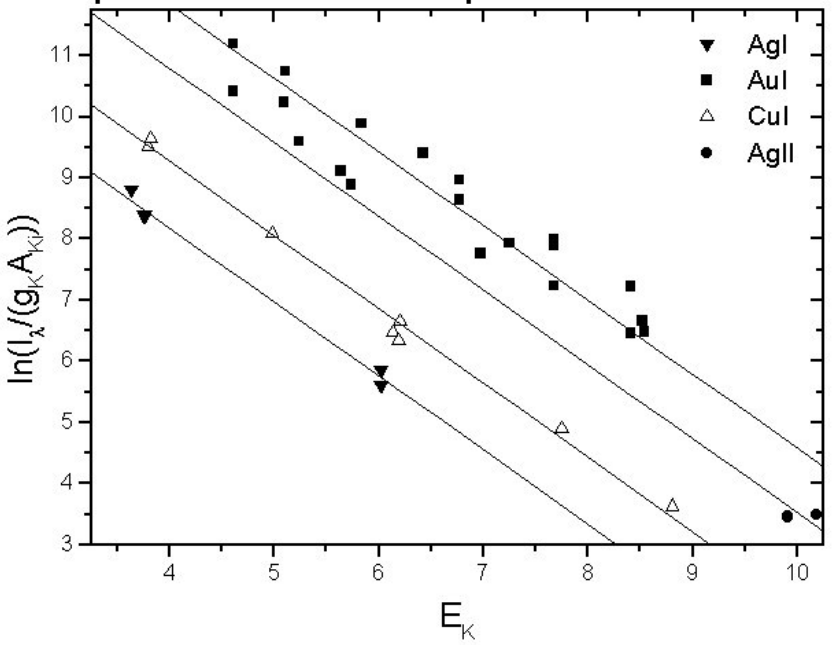
$$m = -\frac{1}{k_B T}$$

$$x = E_k$$

$$q_s = \ln\frac{C_s F}{U_s(T)}$$

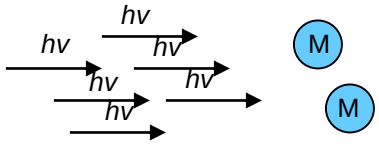


Example of Boltzmann plot of Golden alloy



Plasma Radiation (Emission)

- Laser beam (flux of photons or flux of energy)
  - Free electrons are necessary to start the LIPS process
  - Seed free electrons
    - Can naturally occur (cosmic ray, radioactivity) but low probability
    - Formed by multi-photon ionization



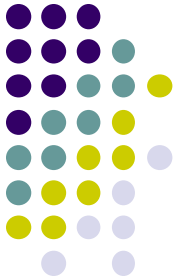
- Multi-photon ionization needs a high photon density.

$M + n(hv) \rightarrow e^- + M^+$

*n depends on the wavelength (energy) of the incident photon

- n = ~8-12 photons for a λ = 1064 nm or ~2-3 photons for a UV source (λ < 400 nm)
- Increase photon density by focusing laser, using very short pulses (~ns), and by increasing laser pulse energy.

Plasma Radiation (Emission) cont.



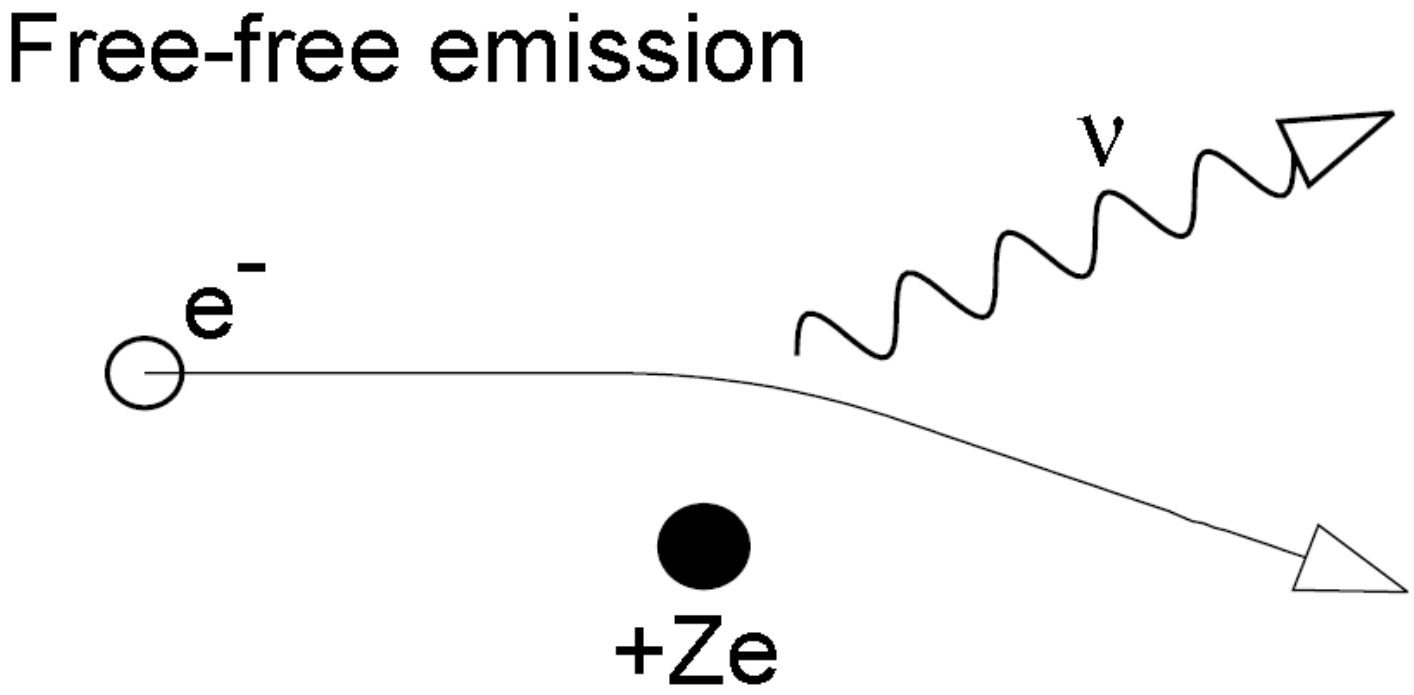
- Once free electrons are present, these electrons absorbing energy from the succeeding photons of the laser beam by inverse Bremsstrahlung. The later produced two lower energy electrons. The energy absorption from the laser is repeated by these 2 e, making ionization and so on. In this manner cascade ionization is developed.

  (1) $e^- + h\nu \rightarrow e^{-*}$ (e^{-*} is a more energetic (KE) free electron)

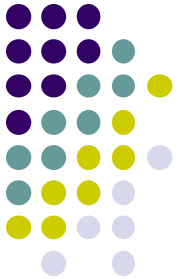
  (2) $e^{-*} + M \rightarrow M^+ + 2e^-$

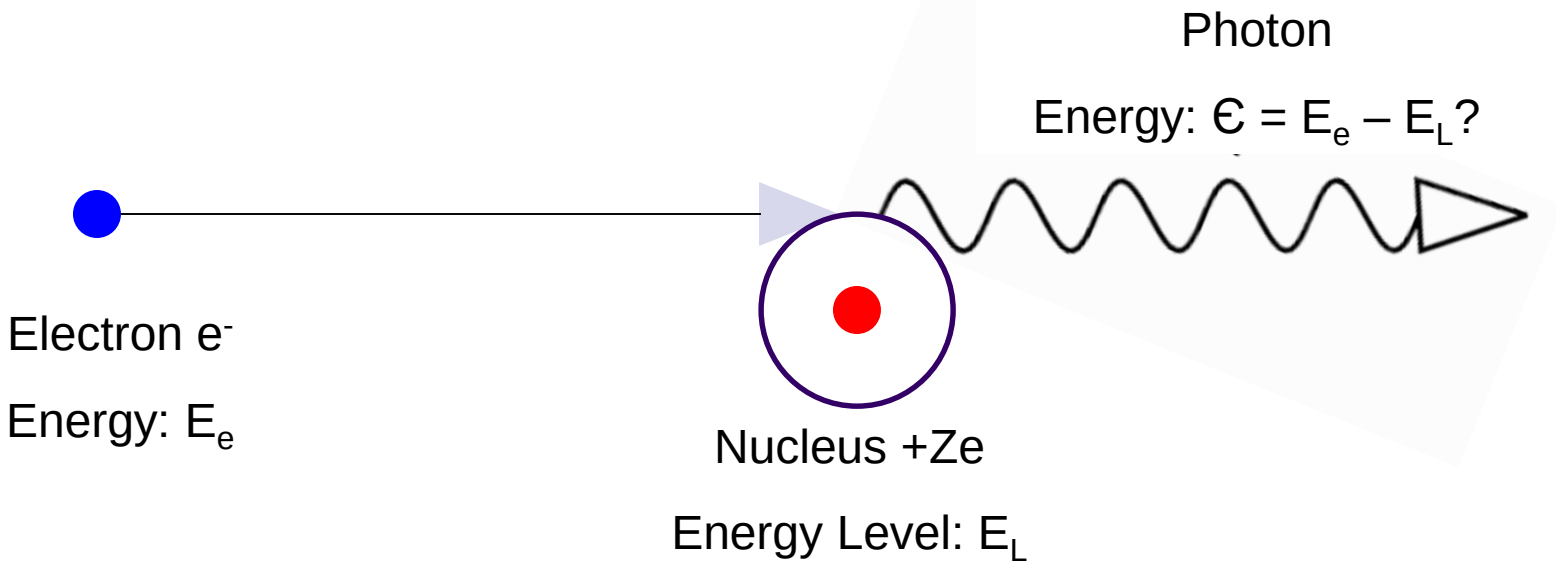
- This process rapidly increases T and N_e.
- By the end of the laser pulse (pulse duration ~10 ns)
  - 50%-90% of the laser pulse energy has been coupled into plasma
  - T~40,000K
  - N_e~10^{18} to 10^{19} #/cc
  - Plasma Volume has expanded to about $1mm^3$

Continuum Emission:
(1) Free-Free Emission
Bremsstrahlung

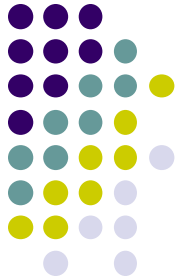


Continuum Emission:
(2) Free-Bound Emission
Recombination Radiation





Plasma Radiation (Emission) cont.



- Early (~ ns & < μs) – High T's and N_e's result in <u>continuum emission</u>
  - Strong broadband emission in the UV and Visible light range that can approach blackbody radiation
  - Sources of continuum emission include:

    Free electron recombination (free-bound) – the free electron will release light when it becomes quantized in an electron shell

    Bremsstrahlung (free-free) - free electron passing by and ion can decelerate causing a KE adjustment resulting in light emission
- Later (~1-30 μs): T's and N_e's decrease.

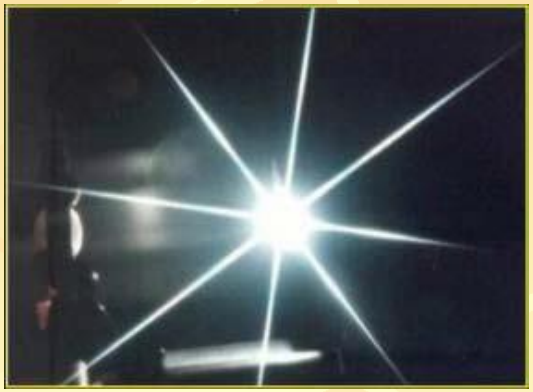
  Free electrons become quantized and relax toward electronic ground states emitting element specific light (unique wavelength) creating atomic emission lines (This is what we measure).

  The optimal time to resolve the atomic emission lines is element specific.
- After ~30-50 μs: Atoms form Molecules which have weak emission bands
- After ~50 μs: All emission is undetectable

Breakdown of gas

When a laser pulse of sufficiently high power (few MW) is focussed by a lens in atmospheric air or in a gas produces the breakdown, accompanied by a bright spark

Multi-photon absorption produces few free electrons



Free electrons colliding with atoms and ions gain energy from the beam absorbing photons in Inverse Bremsstrahlung processes.

$$\frac{dn_e}{dt} = (\nu_i - \nu_d - \nu_c) n_e - \nu_r n_e^2$$

ν_i	**ionization rate diffusion**
ν_d	**rate recombination rate**
ν_r	**inelastic collisions rate**
ν_c	

If diffusion and inelastic collisions rates are low an avalanche of electrons develops producing an hot plasma

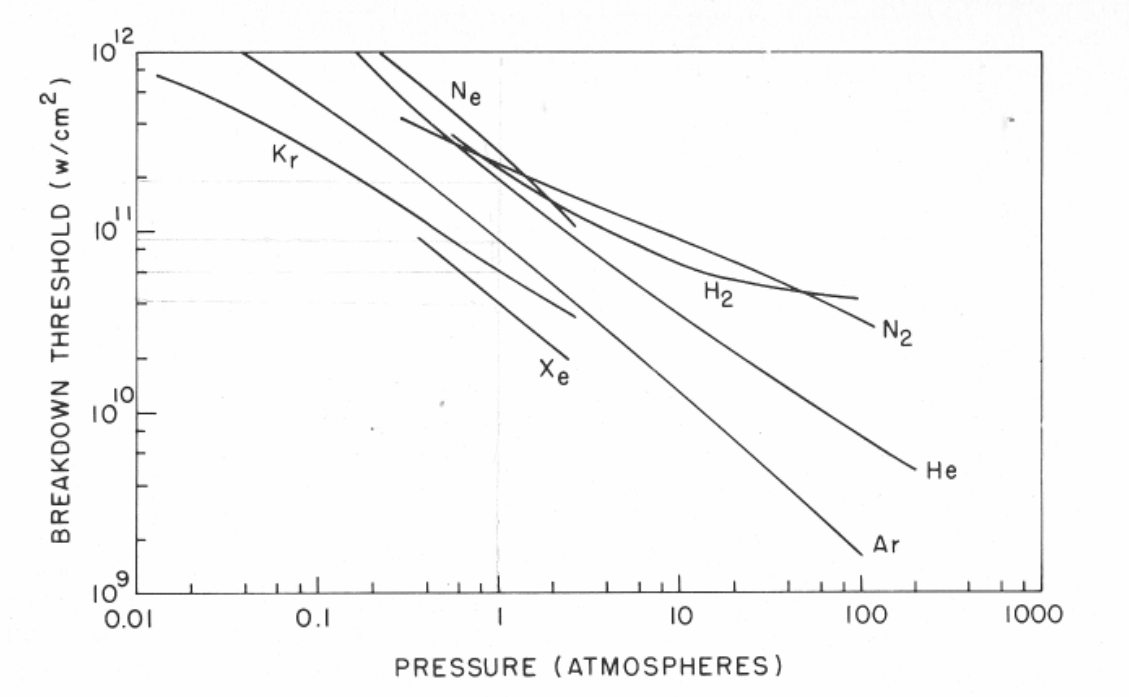
$$n_e(t) = n_{e0} \exp[(\nu_i - \nu_d - \nu_c)t]$$

Breakdown threshold

The electron-ion IB process is much more effective than electron-atom IB process

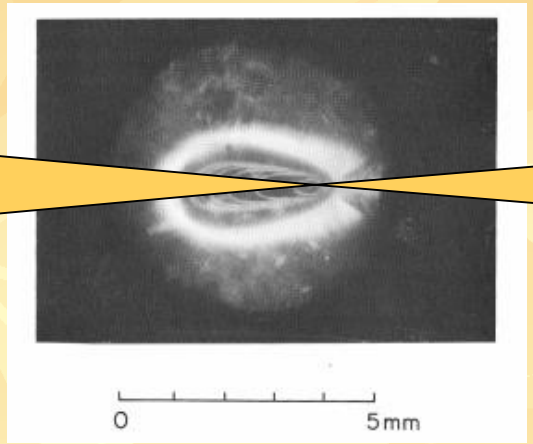
So, the breakdown threshold is defined by the attainement of the ionization degree for which electron-ion IB becomes dominant on electron-atom IB.

Usually is taken $n_e/n_a = 10^{-3}$



the shock wave

Laser pulse



Schlieren photograph of laser spark in Ne at pressure 1000 Torr produced by a ruby laser

The plasma expands producing a shock wave

1stage: Laser Detonation Wave, until the end of the laser pulse

2stage: Blast Wave, during the early microseconds of plasma expansion, strong explosion theory

3stage: Force Drag model, viscous force by buffer gas

Absorption of the radiation and heating of a solid target

Light is absorbed by interaction with electrons ⇒ **Absorption**

The excited electrons collide with lattice phonons and with other electrons and give up their energy ⇒ **Heating**

$\tau_{coll} \approx 10^{-13}\,\mathrm{sec}$ ⇒ **In times of the order of the duration of laser pulse the electrons will make many collisions**

⇓

1. **We can regard the optical energy as being instantaneously turned into heat**
   2.A local equilibrium is rapidly established during the pulse
   3.The concept of temperature is valid and we can use the equations for heat flow

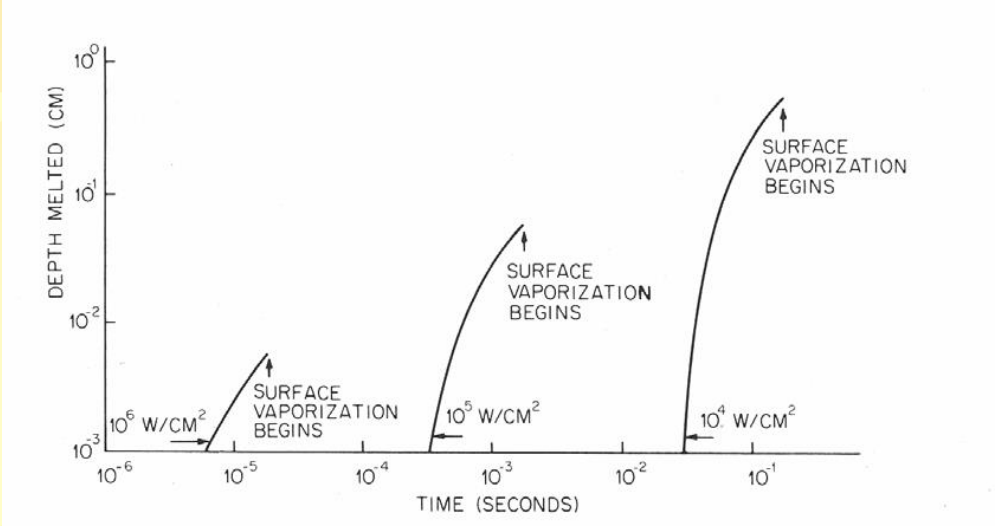
This assumption can break down for the case of subnanoseconds pulse

Melting

When the surface reaches the temperature of fusion, the sample starts to melt.

It is important for welding applications: in this case we want to obtain melting without vaporization, there is just a little range of flux densities suitable for this.
We need not too short and energetic pulses
normal pulse or continuous lasers are used

Element	Heat of fusion (cal mole-1)	Heat of vaporization (cal mole-1)
Cu	2550	19370
Al	3120	21390
Fe	3670	32210
Pb	1141	12830



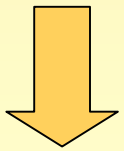
Vaporization

When the temperature reaches the boiling point the surface vaporizes

Vaporization is the dominant process for material removal for irradiance of about 10^6-10^7 W/cm^2. This range is generally obtained with microsecond pulses or longer

Vaporization is a thermal process and is strongly affected by the thermal properties of the target

Elements with lower vaporization temperature will be enriched in the vapor relative to their concentration in the solid

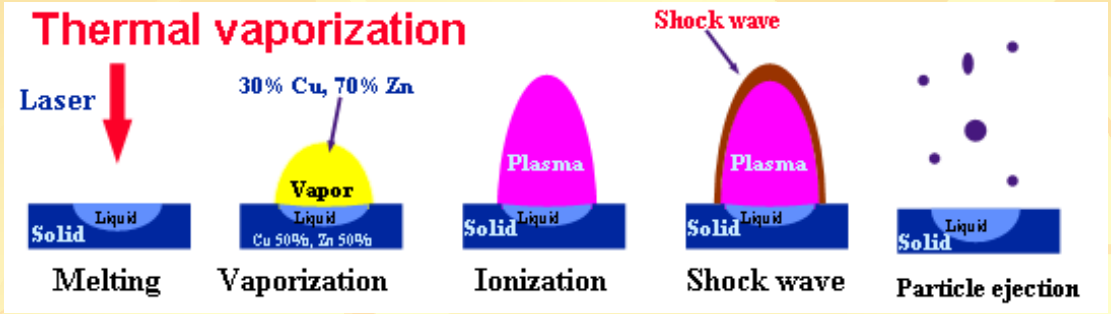


The sampling is not stoichiometric !

Vaporization or Ablation ?

Irradiance of 10^6-10^7 W/cm^2

- thermal process
- melting, vaporization
- non stoichiometric
- mass removal : few milligrams
- crater depth : few millimeters



Irradiances higher than 10^9 W/cm^2

- explosion, non thermal, stoichiometric
- melting and vaporization minoritary
- mass removal: few nanograms
- crater depth: few microns
- laser shielding
- plasma formation
- shock wave

Ablation

At irradiances higher than 10^9 W/cm^2, obtained with nanosecond pulses focused onto any material, an explosion occurs.

Multi-photon absorption

One-photon absorption

Dielectric breakdown

The surface temperature grows very rapidly exceeding the vaporization point within a fraction of the laser pulse

Before the surface layer can vaporize the underlying material will reach its vaporization temperature, increasing temperature and pressure.

The surface explodes

The processes involved are predominantly non-thermal (e.g. melting not always is present)

Ablation is stoichiometric

Laser shielding

The hot blowoff material can absorb the incoming laser radiation

The plasma becomes thermally ionized and opaque for the radiation.
Inverse Bremsstrahlung and photoionization processes absorb energy from the beam

The target is shielded from the plasma and a relatively small fraction of the beam reaches the surface

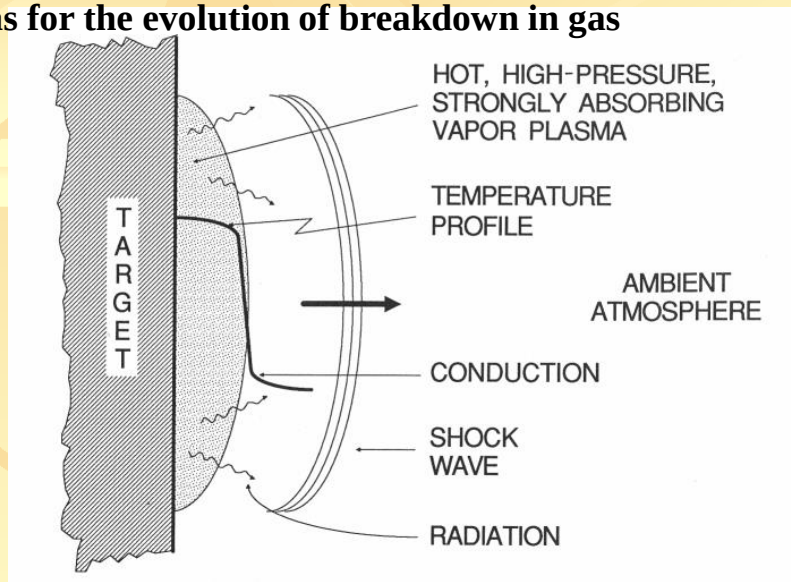
Near the end of the laser pulse, the plasma becomes so hot that it begins to reradiate thermally; this radiation may reach the surface and cause further vaporization

1. **In this way the vapor becomes an hot plasma and expands in the medium**

2. **A given amount of energy delivered at very high power is less effective in removing material than the same amount of energy delivered in a longer, lower-power pulse**

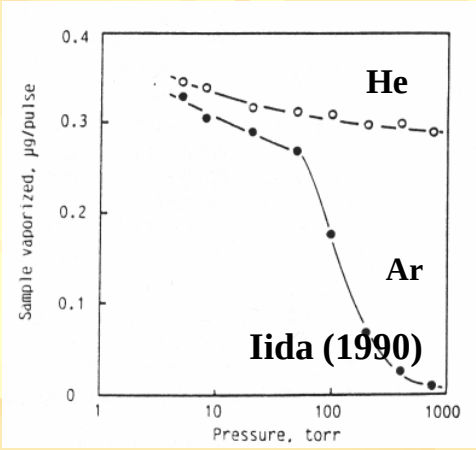
Effect of ambient conditions

When the plasma is induced in a background gas also its molecules are heated, ionized and absorb energy from the laser beam
The plasma expands producing a shock wave
as for the evolution of breakdown in gas



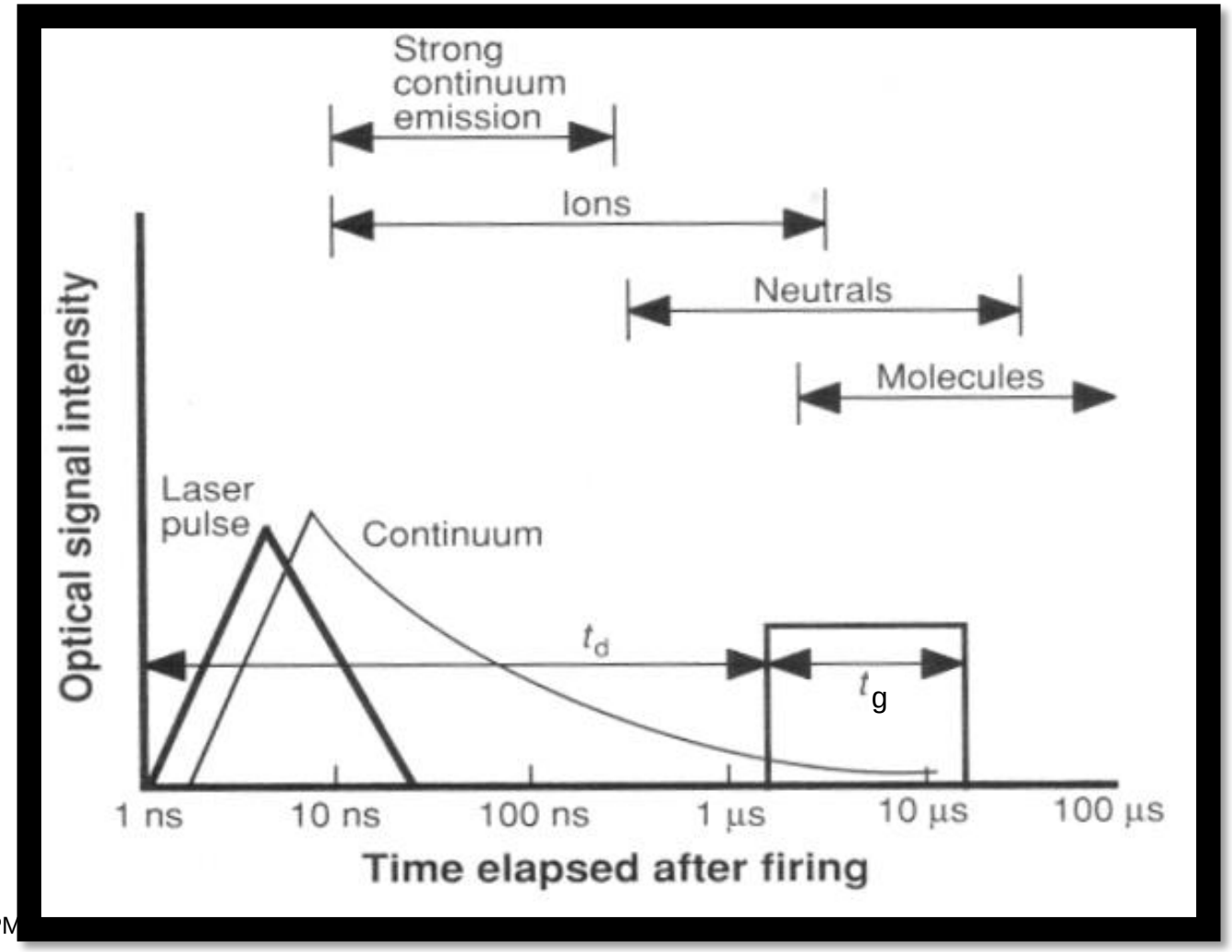
So the composition and pressure of buffer gas affects:

- **Laser shielding and quantity of sample removed**
- **Temperature and electron density of the plasma**
- **Expansion of the plasma**
- **Emission of the plasma**

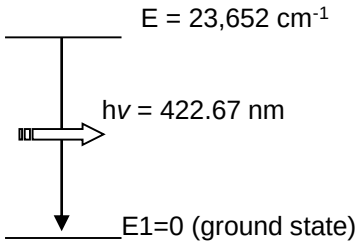


Al sample, 120 mJ, $3.8 \cdot 10^{10}$ W/cm^2·sec
100 % absorption ⇨ 8 μg

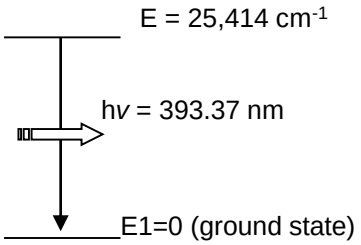
Emission and Collection Timing



Atomic Emission Characteristics



Ca(I) – neutral calcium emission



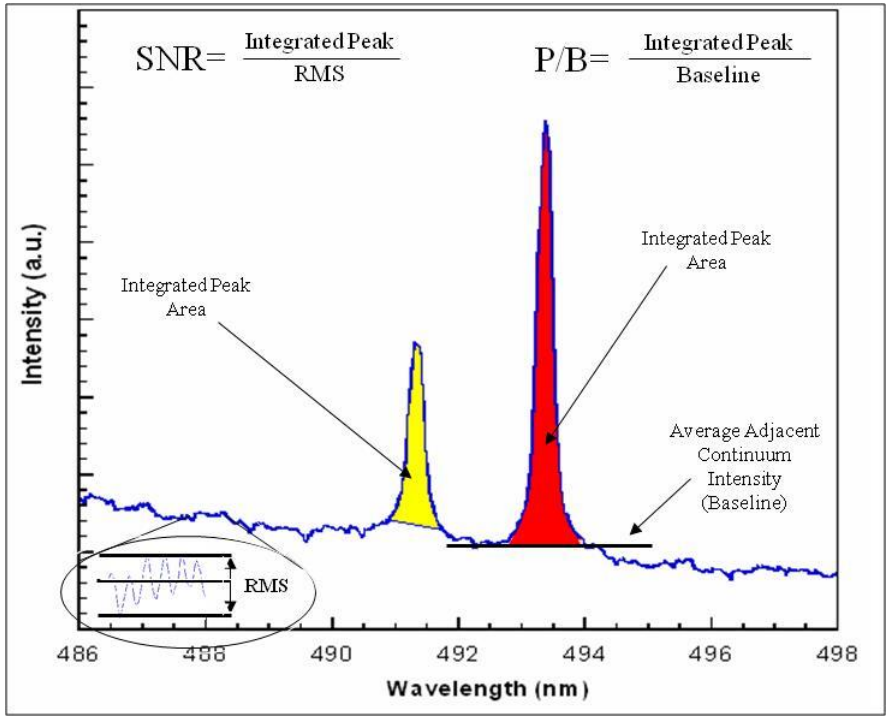
Ca(II) – Ionized calcium emission

- Ions and neutral atoms have different emission lines.
- Atomic emission is proportional to the number of emitting atoms/ions.
  - Allows for concentration measurements

Qualitative LIBS

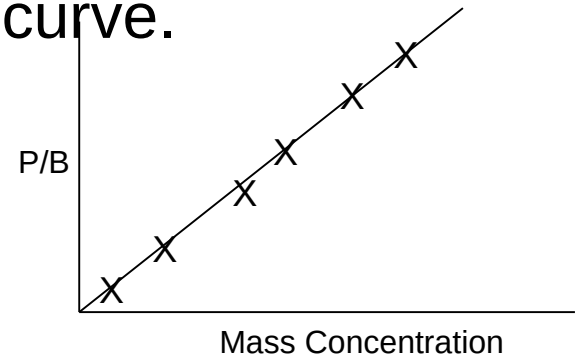
- Measure elemental composition of solids, liquids, gases, and aerosols.

  Compare the wavelengths of detected lines to databases (NIST)
- Measured signal varies with concentration
  - Peak-to-base (P/B) and the Signal-to-noise (SNR).



Qualitative LIBS

- Must Calibrate system
  - Analyze known quantities of analyte and construct a calibration curve.



- Use standards that match the bulk material of the sample of interest (match the matrix)

LOD Limit of Detection

- As defined by **IUPAC**, "International Union of Pure and Applied Chemistry " the detection limit is the concentration required to produce a net line intensity equivalent to three times the standard deviation of the background signal.

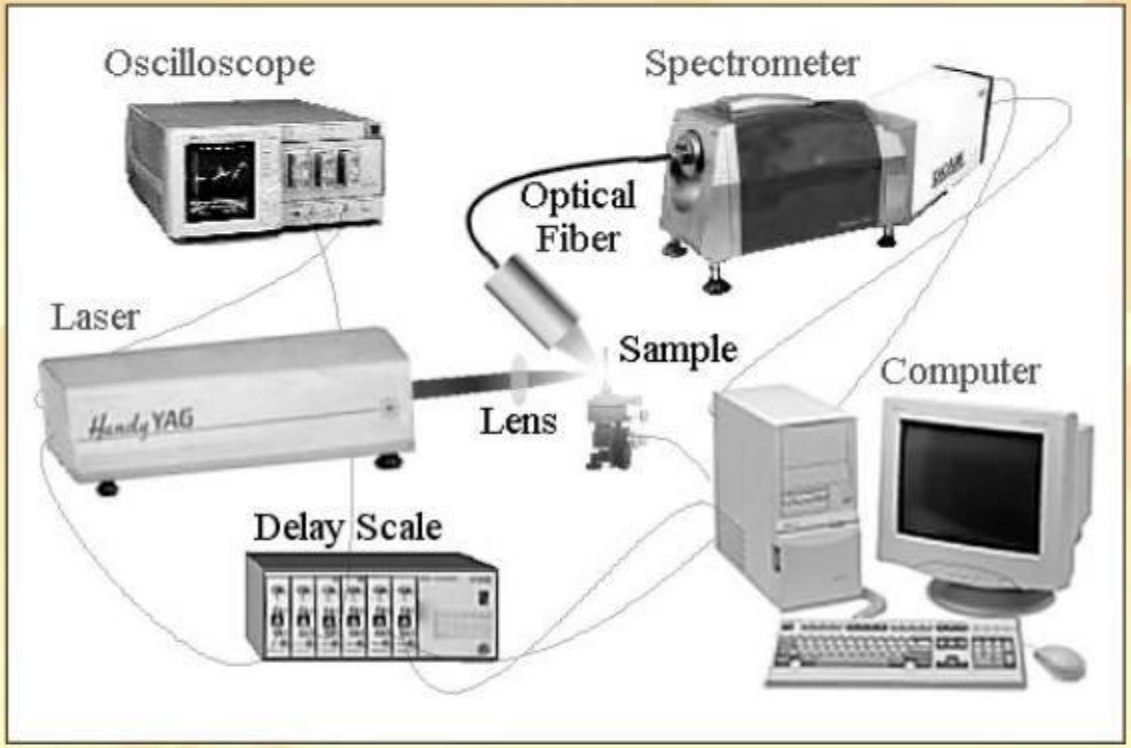
- For such LIPSwe chose the strongest line for each element and based the calculation on the 3 σ IUPAC definitions. The limit of detection was calculated from the formula

$$LOD = 3\,\sigma / s$$

Where σ is the standard deviation of the background and s is the calibration slope.

typical LIBS apparatus

Laser Induced Plasma Spectroscopy (LIPS)
Laser Induced Breakdown Spectroscopy (LIBS)
Laser Ablation Spectroscopy (LAS)



Advantages of LIPS technique

- Micro-destructive technique
- No pre-treatment of sample
- Multielemental Analysis
- High spatial resolution
- Short measurements times
- Instrumentation not much expensive
- Suitable for *in situ* measurements, remote control
- Stratigraphic Analysis

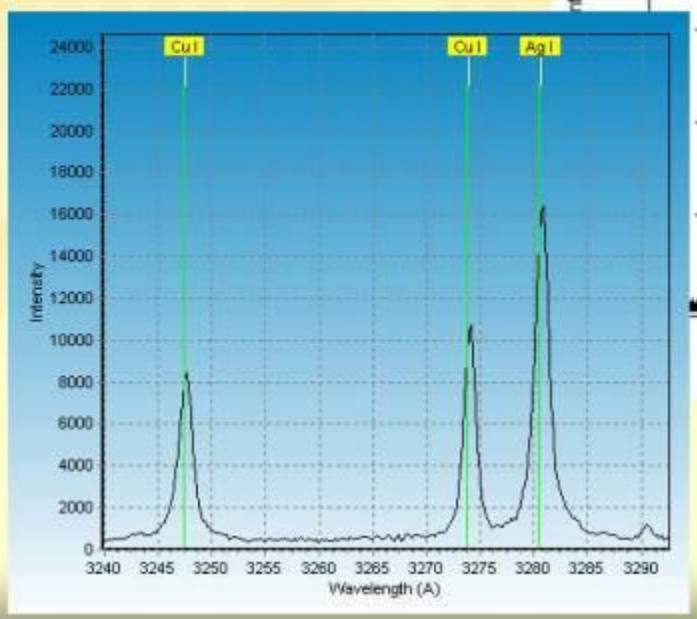
Drawbacks

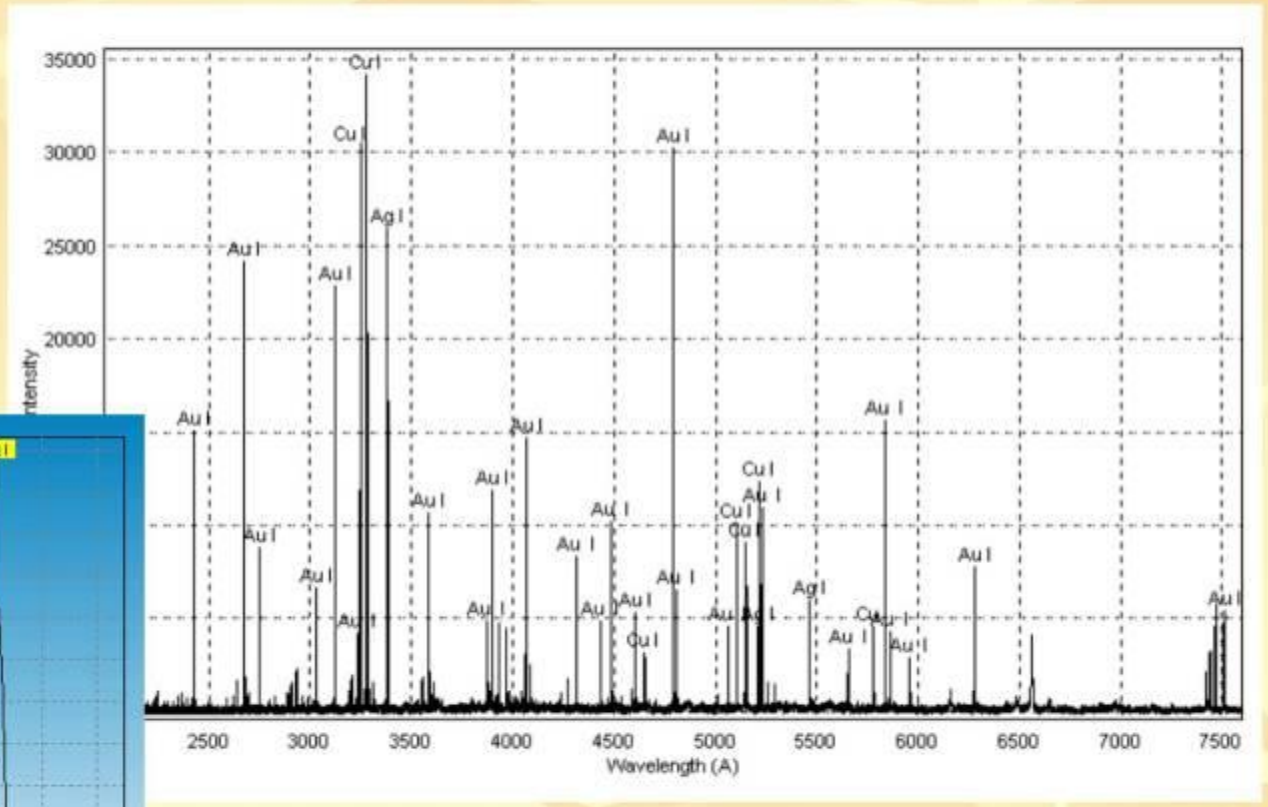
- Detection limits
- Precision and accuracy
- Some elements are hardly detectable (Cl, S, …)

typical LIPS spectrum

(acquired with an echelle spectrometer)

Delay time : 2 μs
Gate time : 1 μs

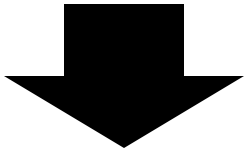




Ternary Alloy : Au, Cu, Ag

LIBS SIGNAL ENHANCEMENT

- Our aim is to enhance S/N ratio by discriminating analyte atomic emission from continuum background emission limits the analysis



a decrease in the value of LOD (Limit of detection)

How to optimize LIBS?

1. **Minimize the Matrix effects**
2. **Avoid Self-absorption**
3. **The internal standardization**
4. **Effect of line choice on linearity**
5. **Effect of Atmospheric Conditions on LIBS Spectra**
6. **Dual pulse LIBS**

1- Minimize the Martix effects

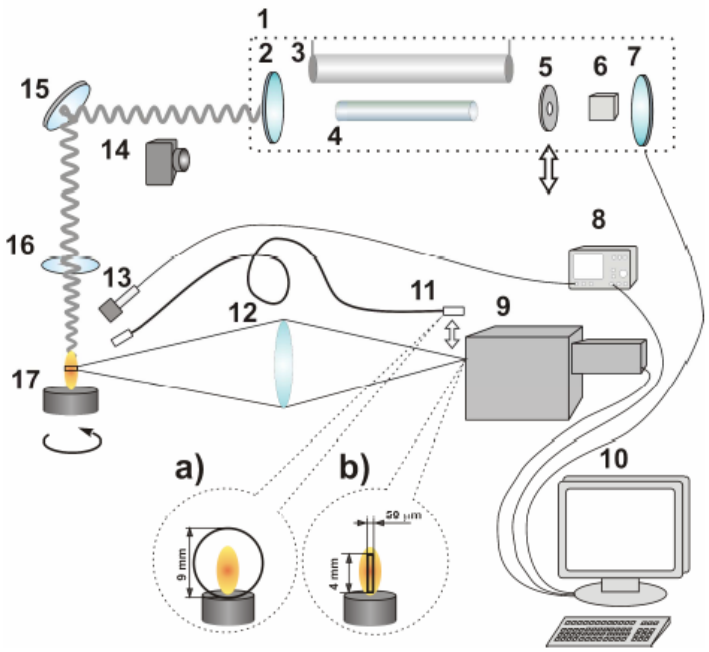
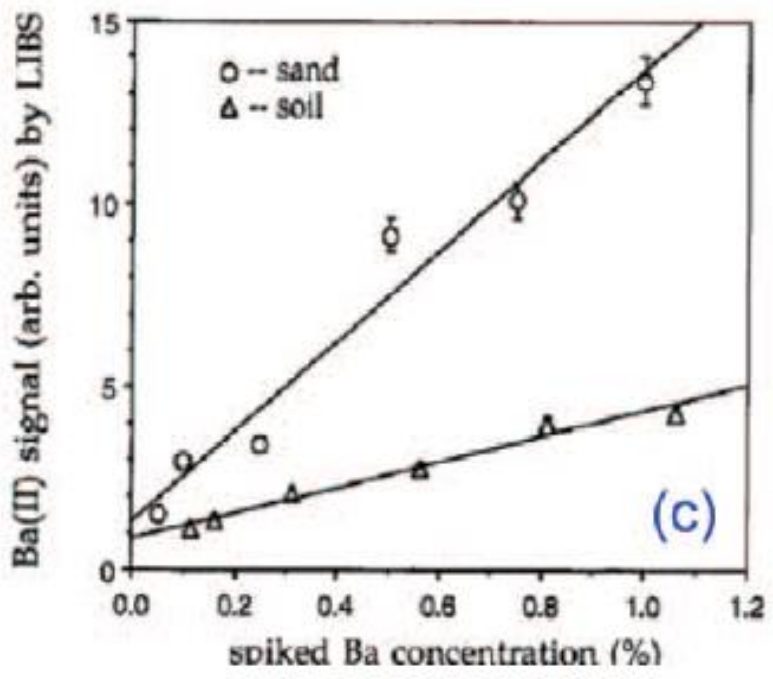


Figure 1. Experimental setup

1. Nd:YAG laser (λ = 1064 nm, 6 mJ/pulse < E < 80 mJ/pulse, τ = 10 ns) 2. front mirror, 3. flash - lamp, 4. active element rod (d=5.9 mm), 5. diaphragm (d=1.4 mm), 6. Q-switch, 7. rear mirror, 8. oscilloscope, 9. spectrograph with ICCD, 10. computer, 11. quartz optical fiber, 12. quartz collecting lens (F = 120 mm), 13. microphone, 14. CMOS camera (for beam profile study), 15. mirror, 16. focusing lens (F = 110 mm), 17. sample (a rotation can be used if needed)

1- Minimize the Martix effects



Matrix effect

Use the same matrix with know elemental concentrations to calibrate your LIBS system !!!

2- the internal standardization (IS) method

To avoid a number of experimental parameters that are difficult to measure, the so-called internal standardization method has been proposed and widely used. The method is based on the following principle: a number of *reference samples* are prepared, all having a similar and known elemental composition. Usually, in such a set of samples, a suitable element dominates (internal standard) and defines the sample properties in view of the 'matrix effects'. It is, therefore, expected that plasma emission be less affected from sample to sample.

In a LIBS experiment, the ratio of the line intensity of a trace element to the emission line of the internal standard is measured and plotted as a function of the known concentration ratios in the reference samples.

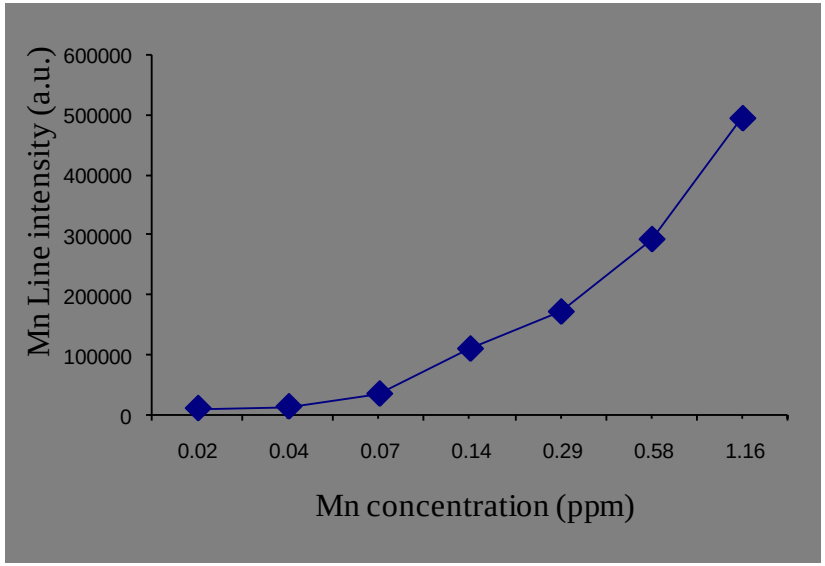
Manganese calibration curve in Al Alloys

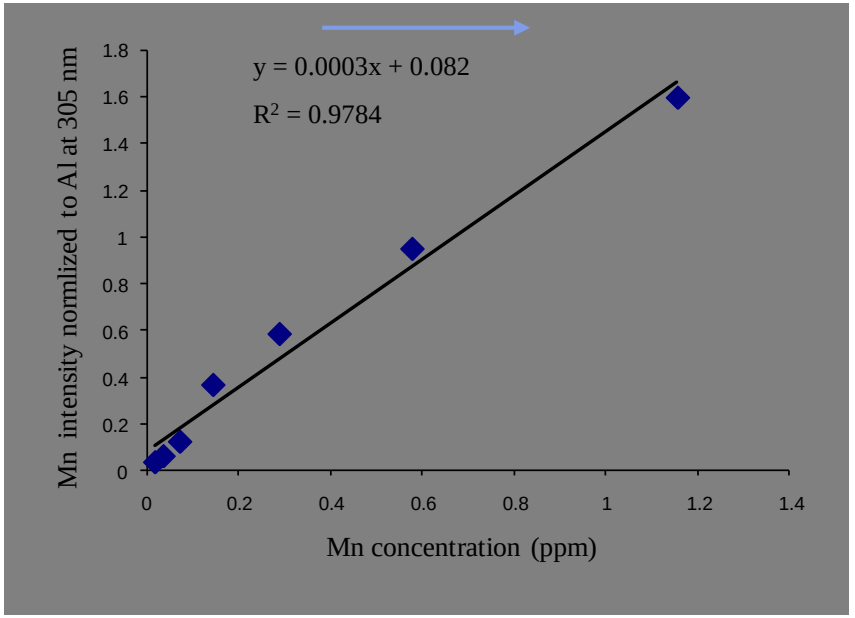
Internal

Standardization

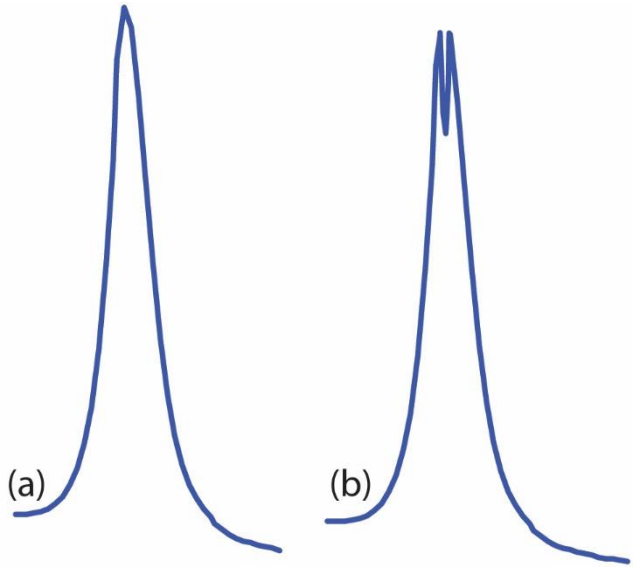
non-normalized

normalized



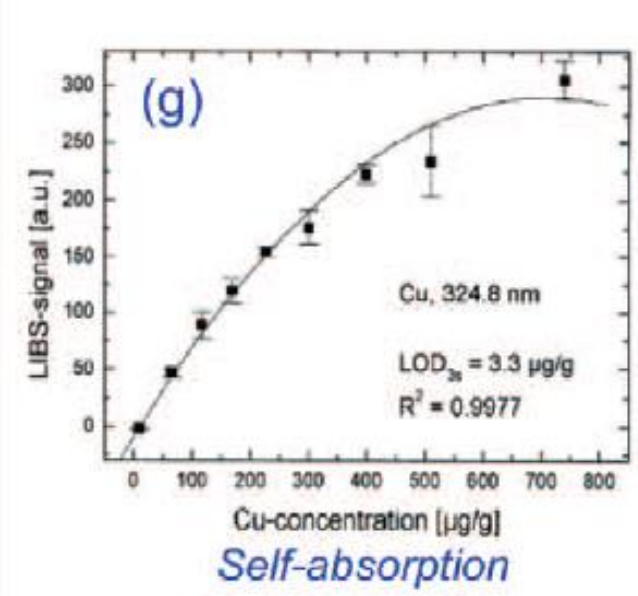


3- avoid Self-absorption (optical thick case)



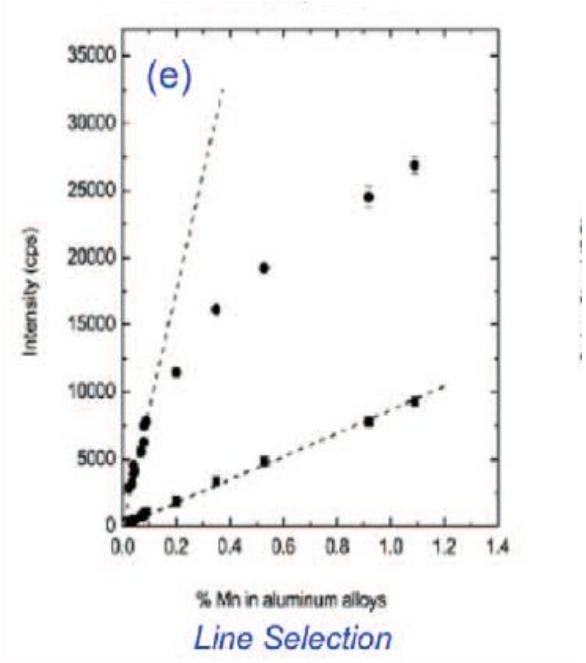
For hot non LTE plasma, two plasmas exist, a primary plasma close to the target surface and a secondary plasma at reduced pressure. The emission from the primary plasma is dominated by continuum radiation while that of the secondary plasma is predominantly characterized by line radiation.

3- avoid Self-absorption (optical thick case)



For hot non LTE plasma, two plasmas exist, a primary plasma close to the target surface and a secondary plasma at reduced pressure. The emission from the primary plasma is dominated by continuum radiation while that of the secondary plasma is predominantly characterized by line radiation.

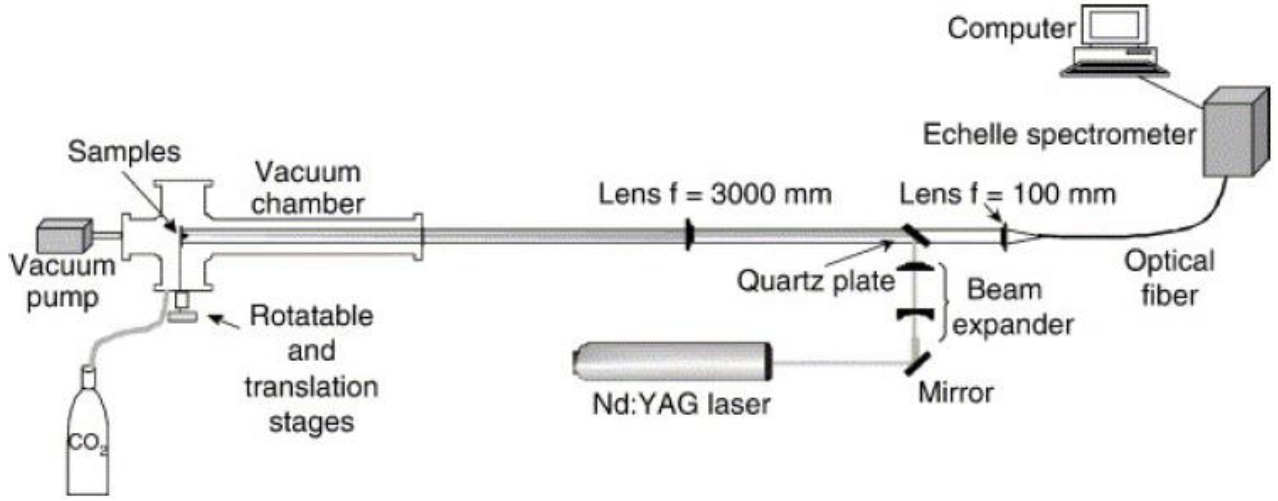
4- Effect of line choice on linearity



avoid interference and selected well isolated lines.

5- Effect of Atmospheric Conditions on LIBS Spectra

Figure 1. Schematic of typical apparatus for a pressure and gas composition LIBS studies. Reprinted from reference [23].

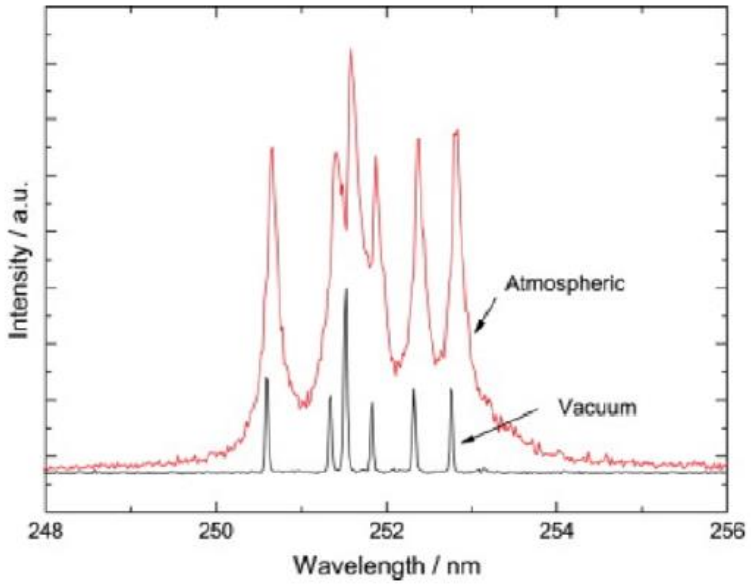


*Sensors **2010**, *10*, 4907-4925

5- Effect of Atmospheric Conditions on LIBS Spectra

1. Low Pressure, <760 Torr

Figure 2. Comparison of LIBS spectra of Si at atmospheric and 10-6 Torr. Reprinted from reference [38].

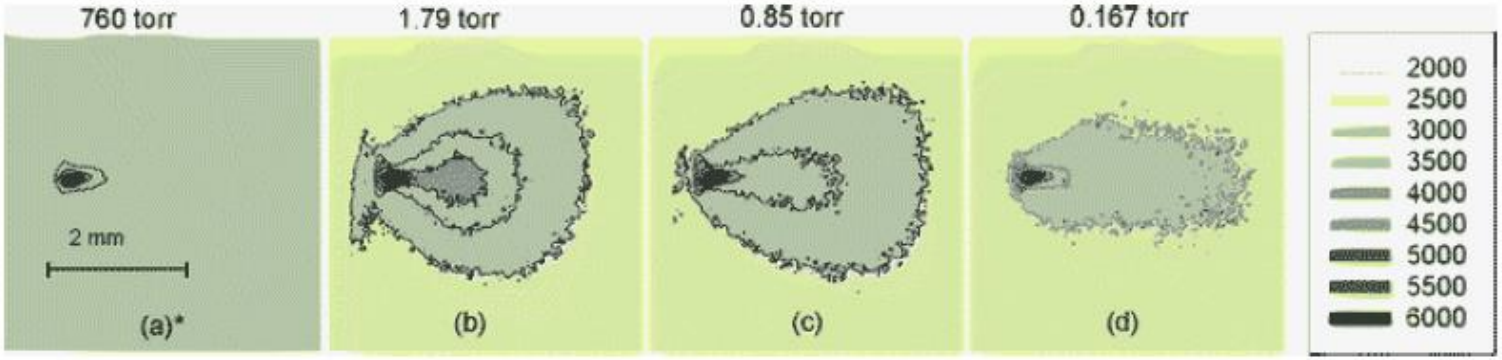


5- Effect of Atmospheric Conditions on LIBS Spectra

1. Low Pressure, <760 Torr

High pressure suppress your plasma

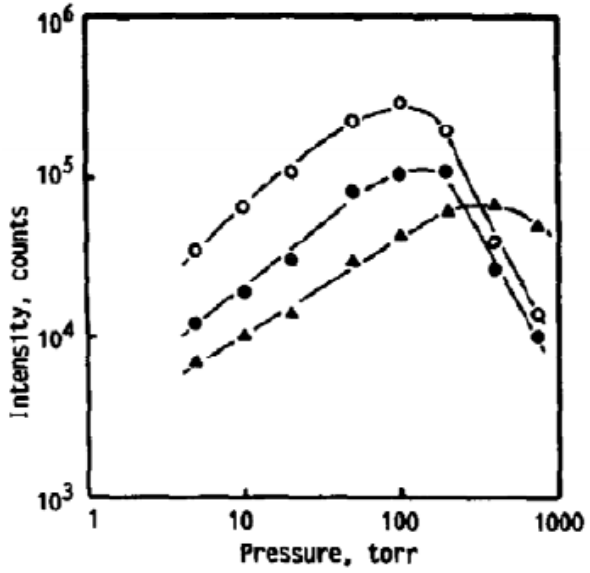
Figure 5. Two-dimensional Cu plasma images at different pressures produced using an accumulation of ten 50 μJ laser pulses, 10 ns second delay and 500 ns gate width. Reprinted from reference [45].



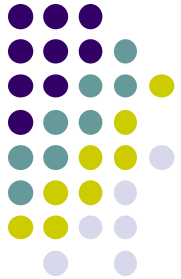
5- Effect of Atmospheric Conditions on LIBS Spectra

2. **Influence of Atmospheric Composition (e.g., He, N2, Ar & CO2)**

Figure 9. LIBS emission intensities of Fe at different atmospheric pressures, Ar (○), air (●), and He (▲). Reprinted from reference [49].



5- Effect of Atmospheric Conditions on LIBS Spectra



2. **Influence of Atmospheric Composition (e.g., He, N2, Ar & CO2)**

Laser plasmas developing rapidly with a sufficient amount of electrons can absorb a significant portion of the laser pulse through inverse Bremsstahlung. Ar is more easily ionized than He and the breakdown threshold in gas for He at 1 atm is ~3 times greater than Ar and ~5 times greater at 100 Torr. Researchers found that the lower breakdown threshold in Ar, compared to He, creates an environment favorable for plasma shielding, and enhance LIBS signal.

5- Effect of Atmospheric Conditions on LIBS Spectra

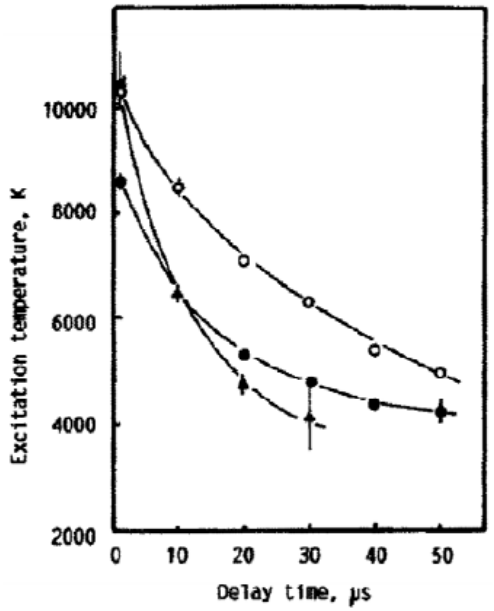
2. **Influence of Atmospheric Composition (e.g., He, N2, Ar & CO2)**

15.759	**Argon**
24.587	**Helium**

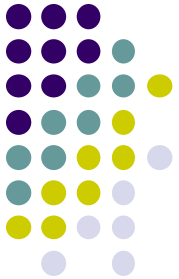
5- Effect of Atmospheric Conditions on LIBS Spectra

3. **Influence of plasma temperature Atmospheric Composition with time**

Figure 10. Temporal resolved plasma temperatures in different atmospheres at 100 Torr, Ar (○), air (●), and He (▲). Reprinted from reference [49].



5- Effect of Atmospheric Conditions on LIBS Spectra



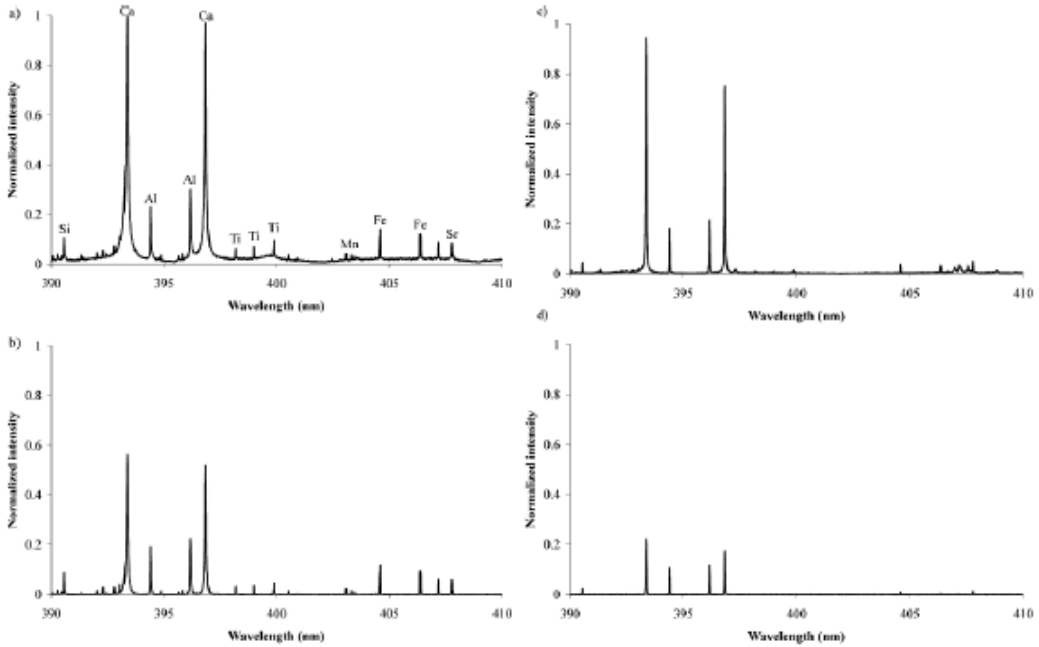
3. **Influence of plasma temperature Atmospheric Composition with time**

It was determined that atmospheric Ar resulted in the highest plasma temperature and electron density, while a He atmosphere resulted in the lowest plasma temperatures and electron density. Studying temporal data, it was also found that Ar had the slowest decay of both electron density and plasma temperature, while He had the fastest decay in both parameters. The reason is the electron density and plasma temperature decay more rapidly in He compared to Ar is because He has a higher thermal conductivity than Ar.

5- Effect of Atmospheric Conditions on LIBS Spectra

3. **Influence of plasma temperature Atmospheric Composition with time**

Figure 13. LIBS spectra of a soil. **(a)** in air no gate delay; **(b)** in air with gate delay of 1 μs; **(c)** in 7 Torr CO_2 with no gate delay; **(d)** in 7 Torr CO_2 with gate delay of 1 μs. Reprinted from reference [15].

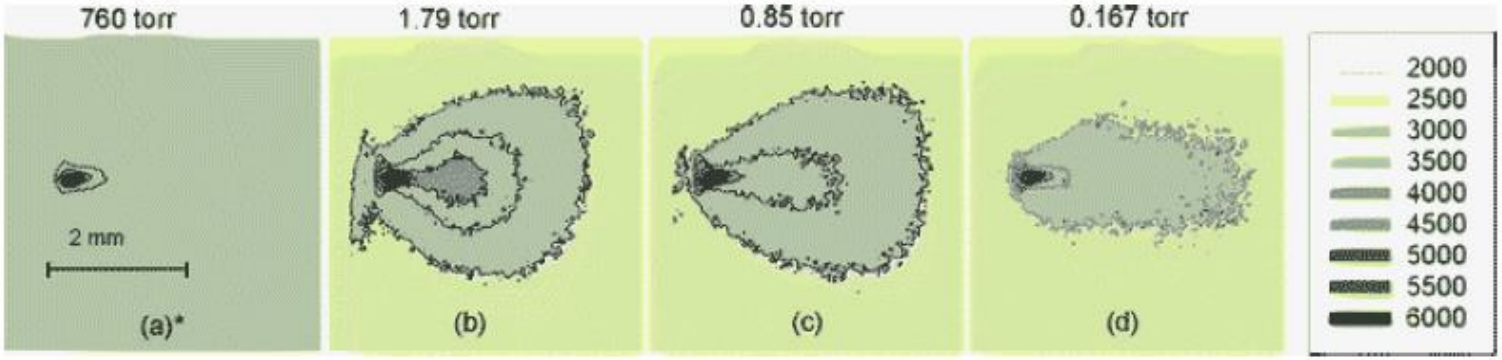


5- Effect of Atmospheric Conditions on LIBS Spectra

1. Low Pressure, <760 Torr

High pressure suppress your plasma

Figure 5. Two-dimensional Cu plasma images at different pressures produced using an accumulation of ten 50 μJ laser pulses, 10 ns second delay and 500 ns gate width. Reprinted from reference [45].



6- Dual pulse LIBS

The rationale behind using double pulse LIBS is the fact that with one single step, the processes of ablation and excitation of the resulting plume cannot be controlled independently.

6- Dual pulse LIBS

How it is work?

Since a large proportion of atoms in the neutral state is present even 40 μs after the plasma formation, a second laser fired at this time could reheat the plasma, thus causing further atom excitation and emission. This increases the emission efficiency so much.

5- Dual pulse LIBS

Solving the matrix effect problem?

The possibility of matrix-independent analysis by selecting time delays at which the intensity ratio of neutral lines with comparable excitation energies are independent of time

6- Dual pulse LIBS

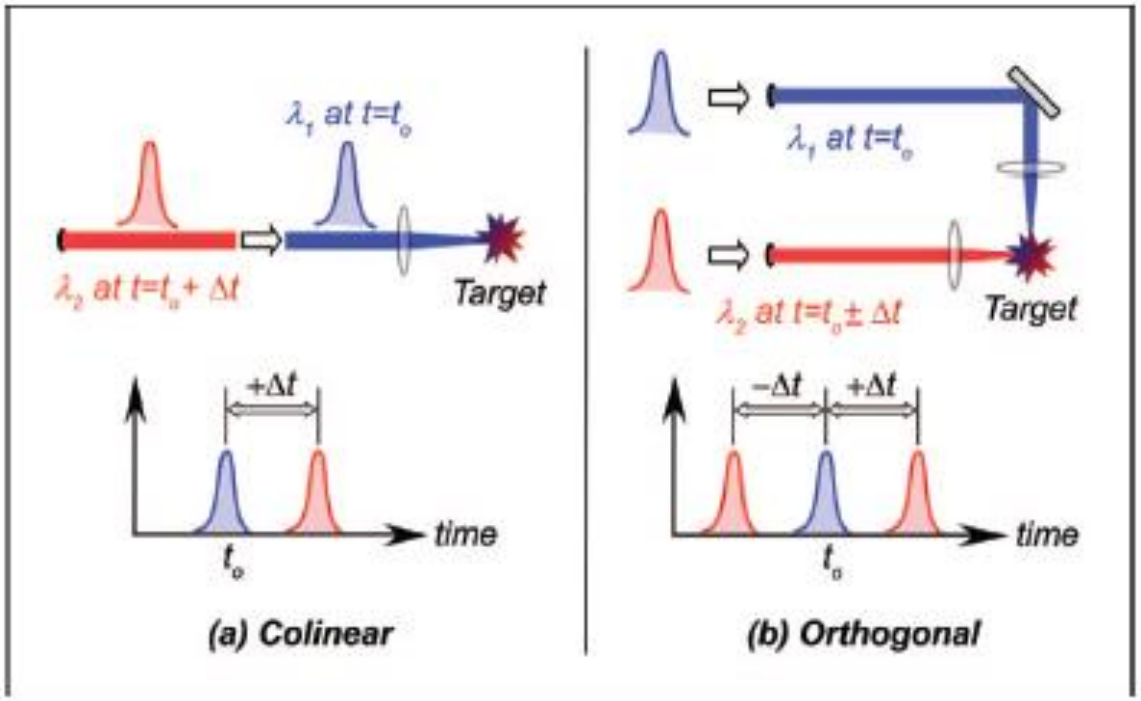
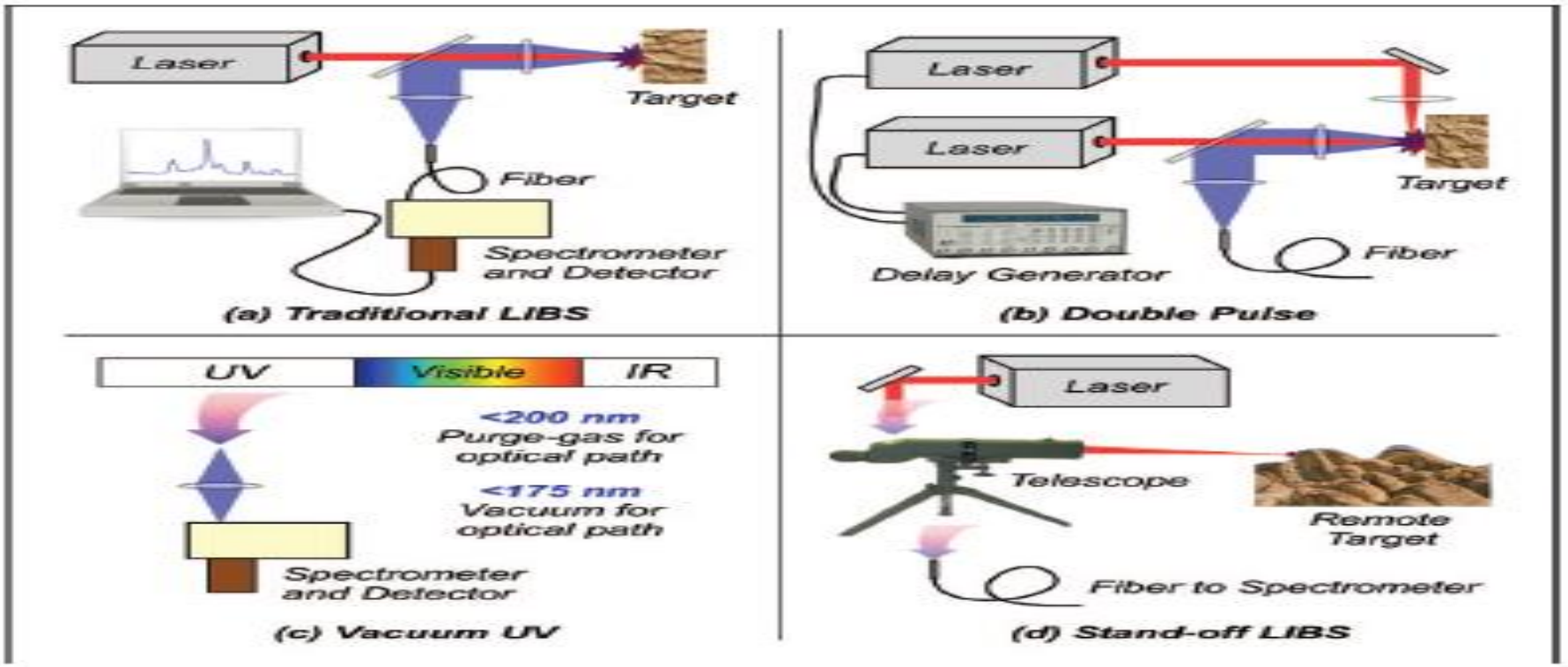
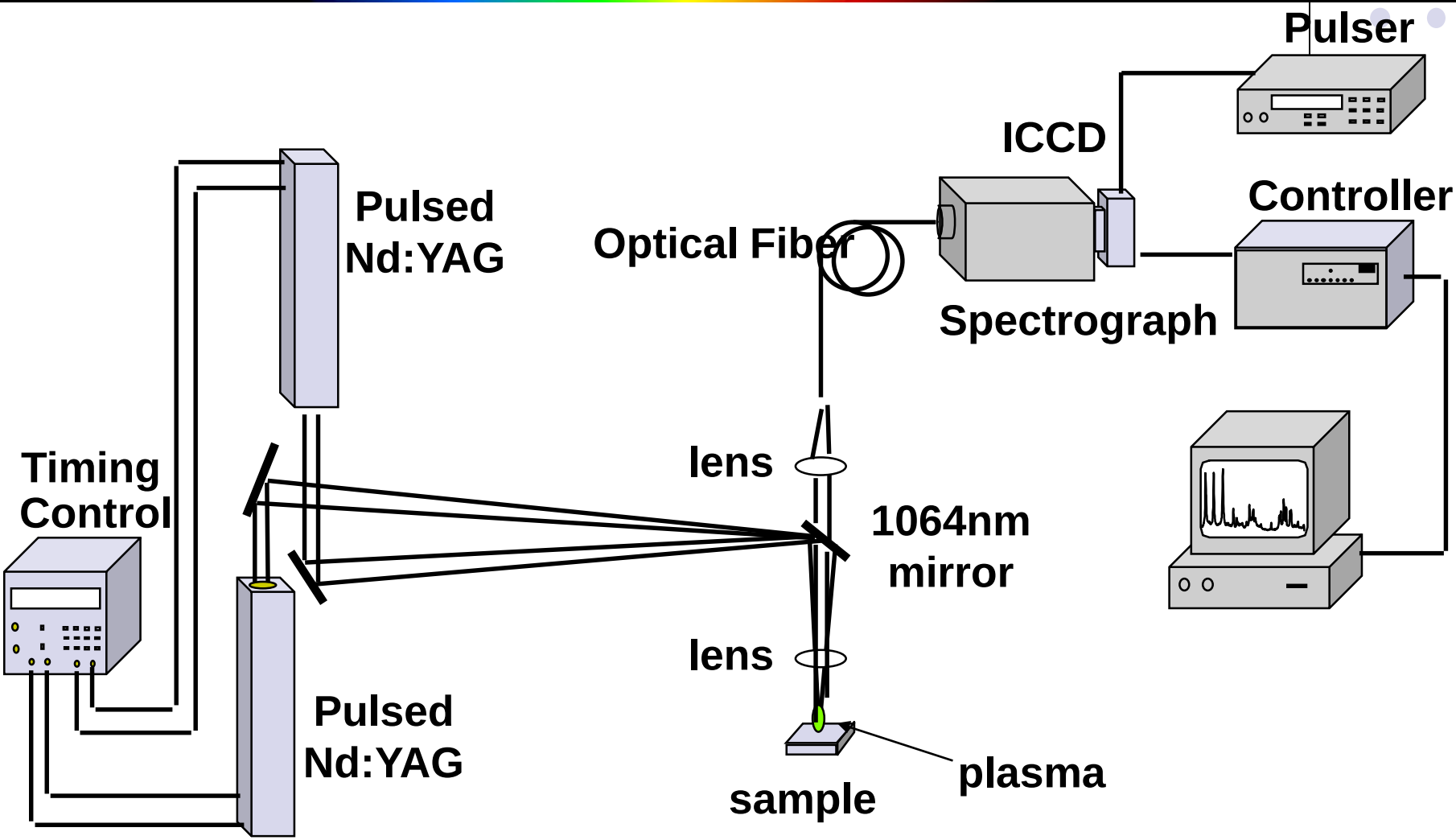


FIG. 9. Schematic illustration of the arrangements used in collinear and orthogonal double-pulse LIBS. In the orthogonal case, the pulse at λ_2 can precede or follow the ablation pulse at λ_1. Note that two wavelengths are shown here for the two lasers.

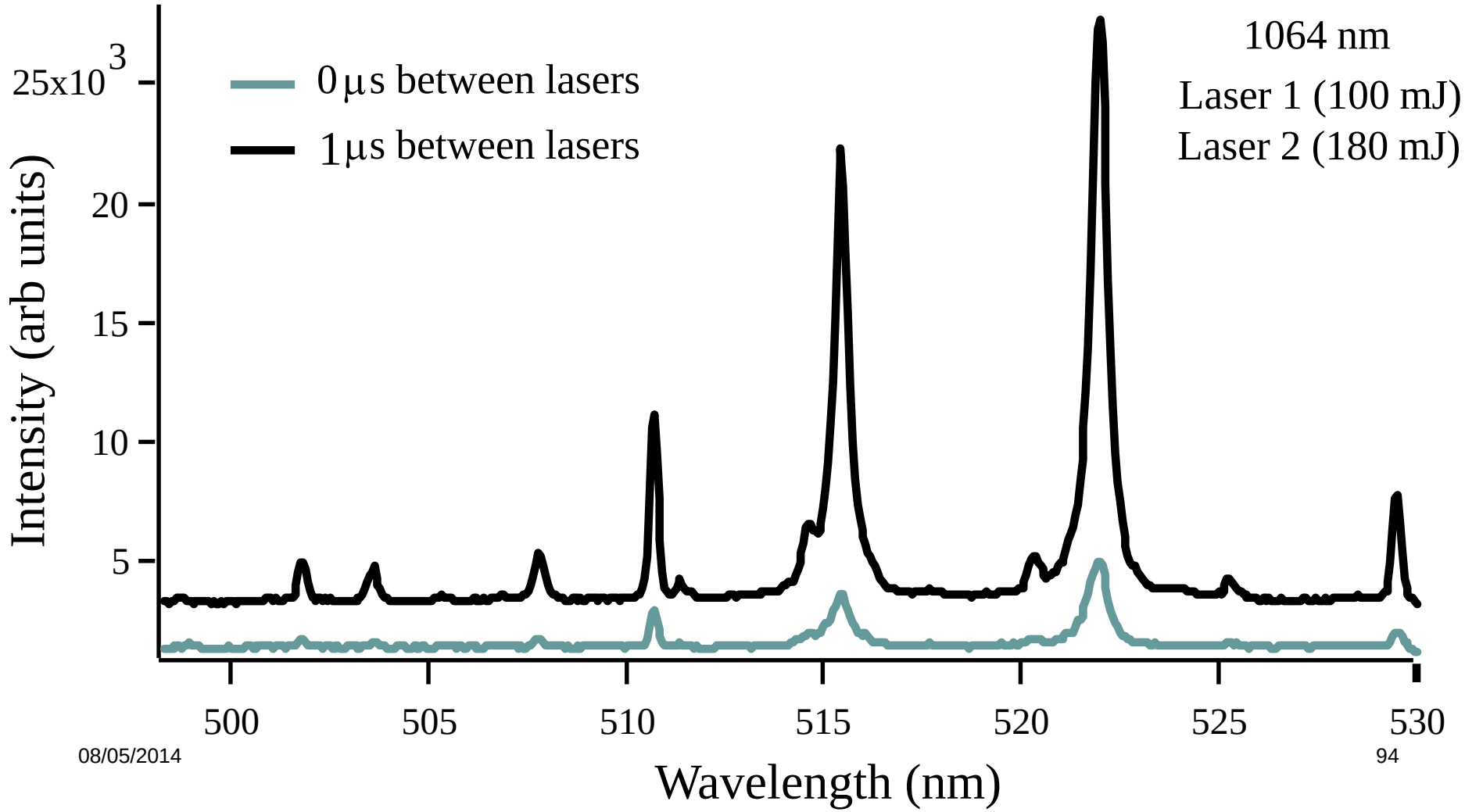
6- Dual pulse LIBS



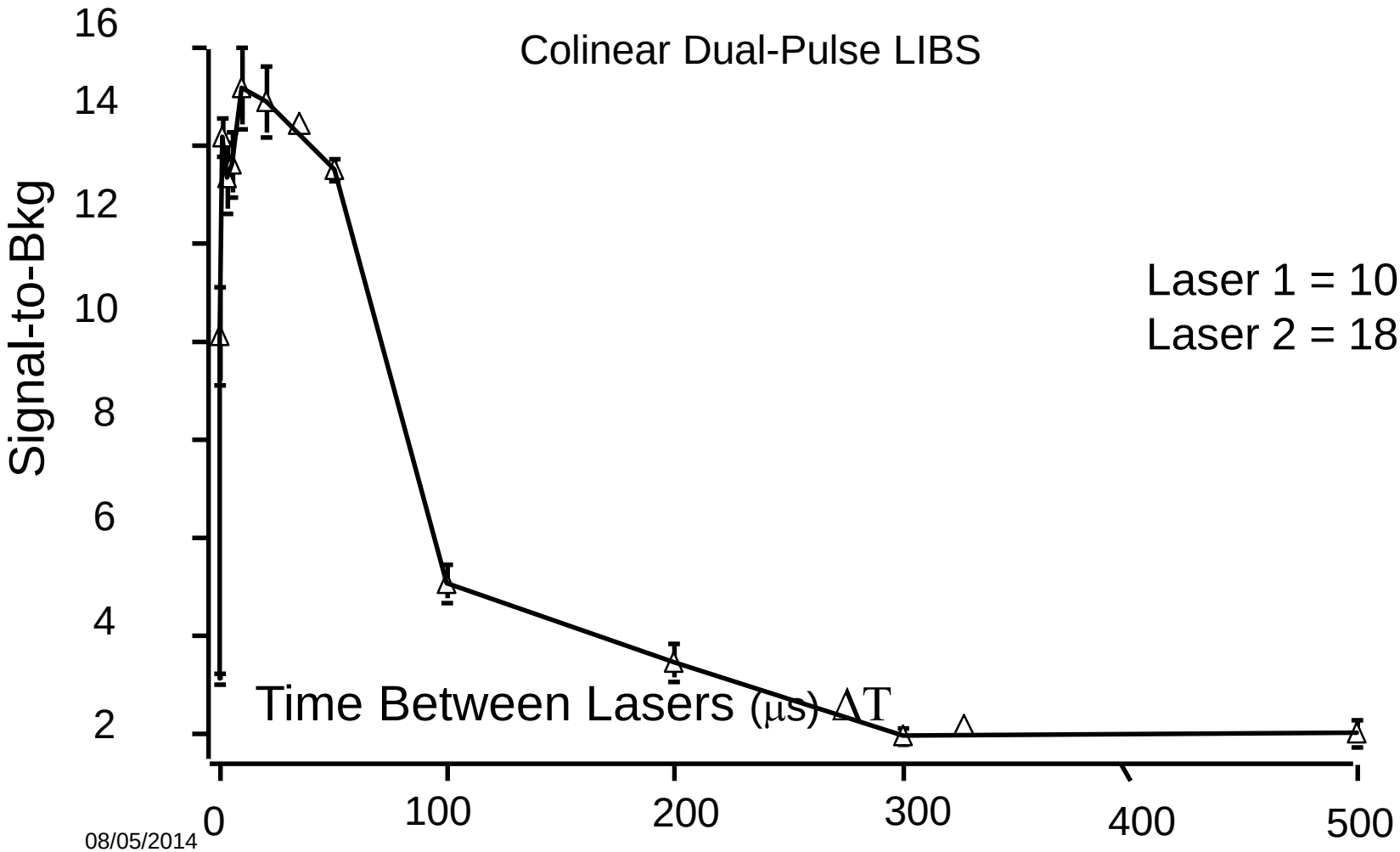
Colinear Dual-Pulse LIBS Configuration



Colinear Dual-Pulse LIBS Enhancement for Copper

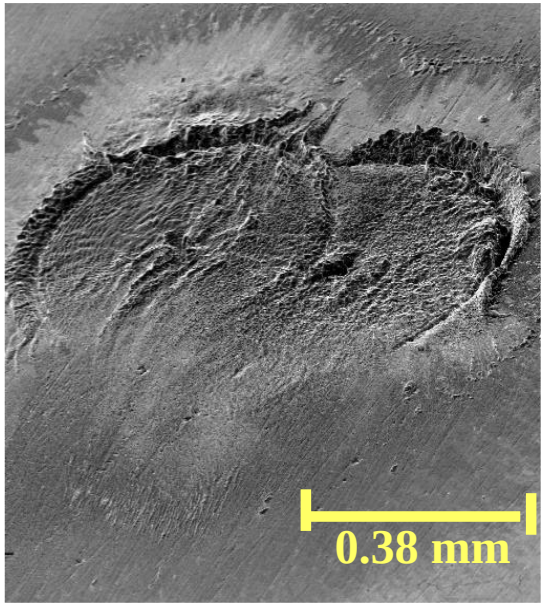


Optimum Delay Between Lasers for Copper Enhancement



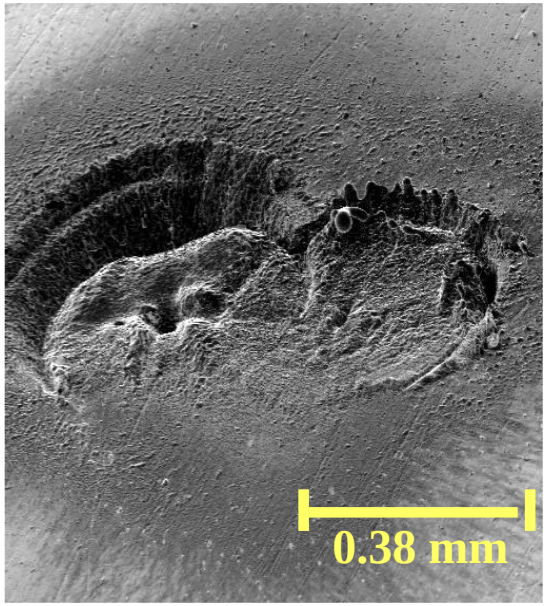
Copper Craters from Colinear Dual-Pulse LIBS

0 μs ΔT



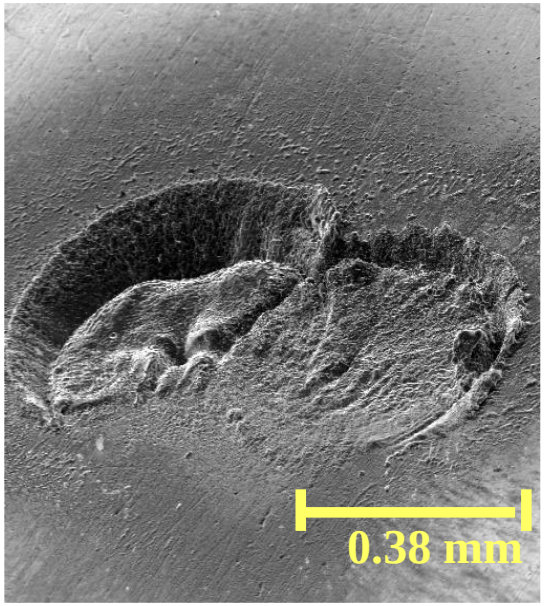
Cu S/B ~ 3

1 μs ΔT



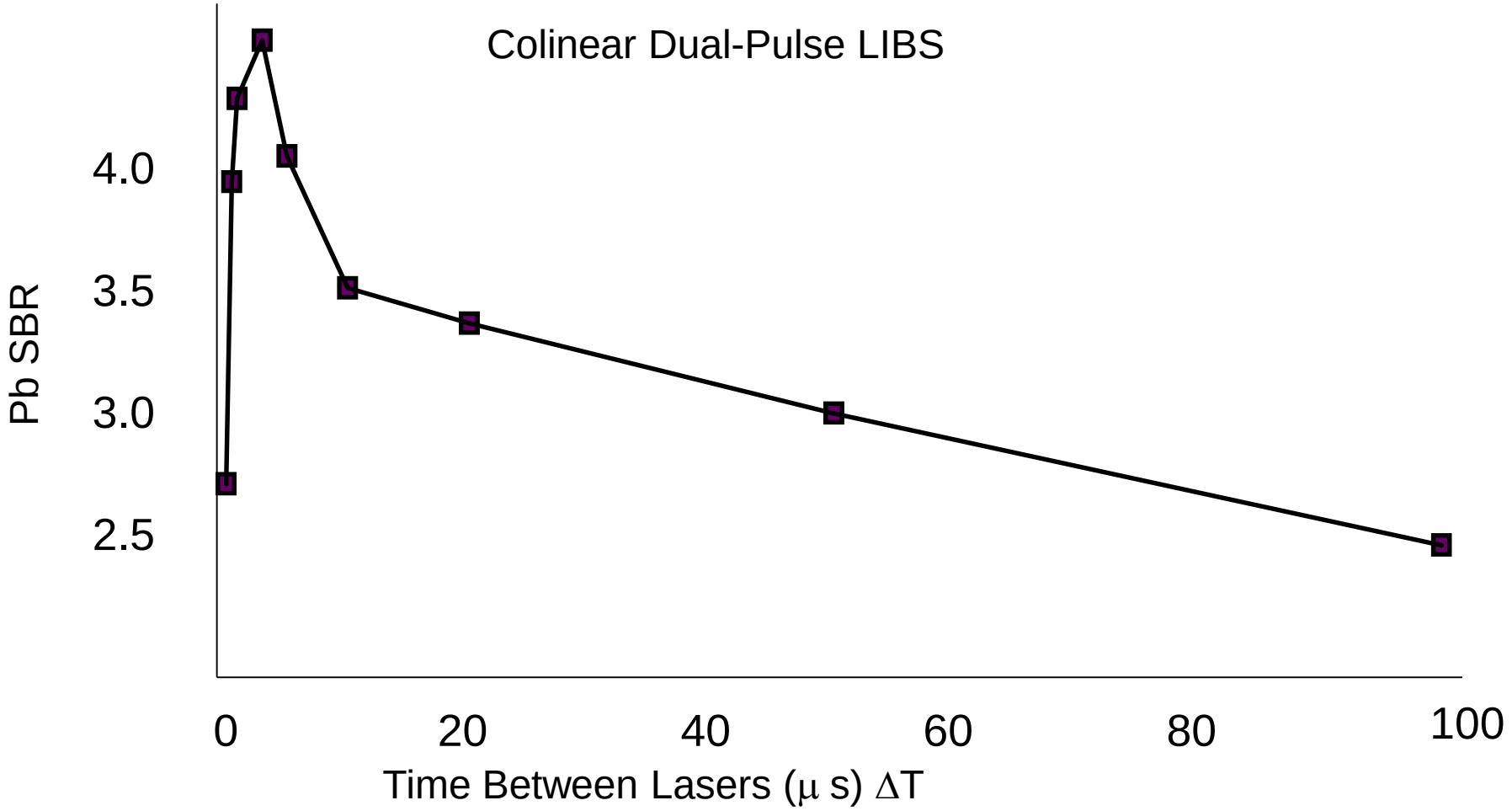
Cu S/B ~ 14

20 μs ΔT



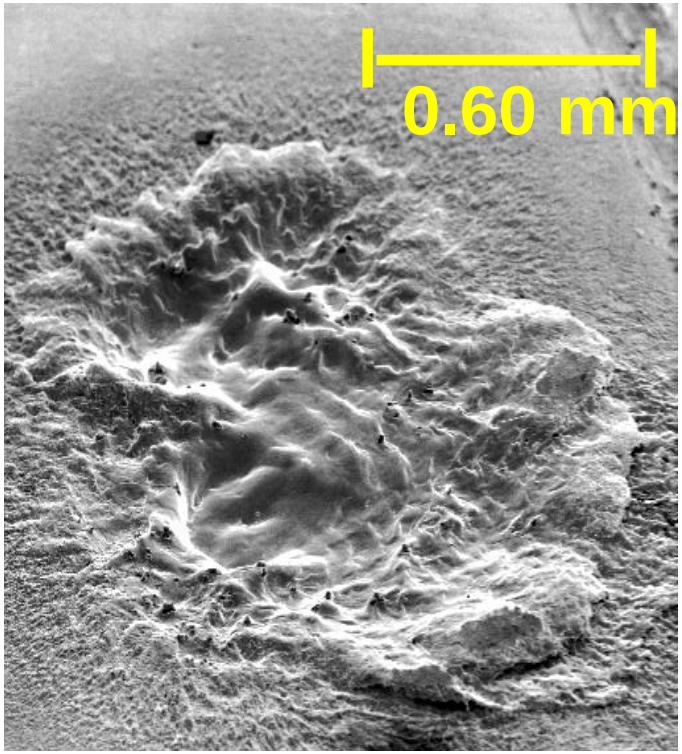
Cu S/B ~ 15

Optimum Timing Between Lasers for Lead Enhancement



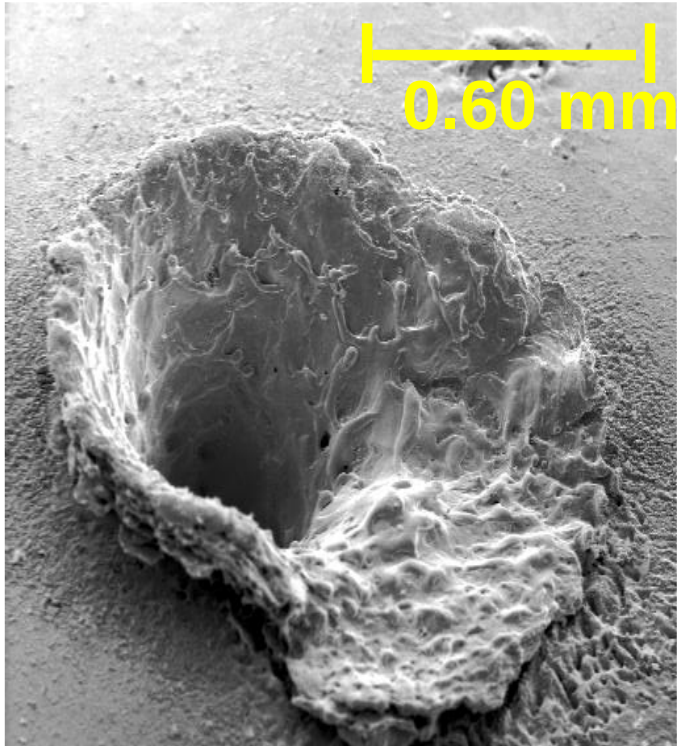
Comparison of Lead Craters (colinear geometry)

Zero μs ΔT



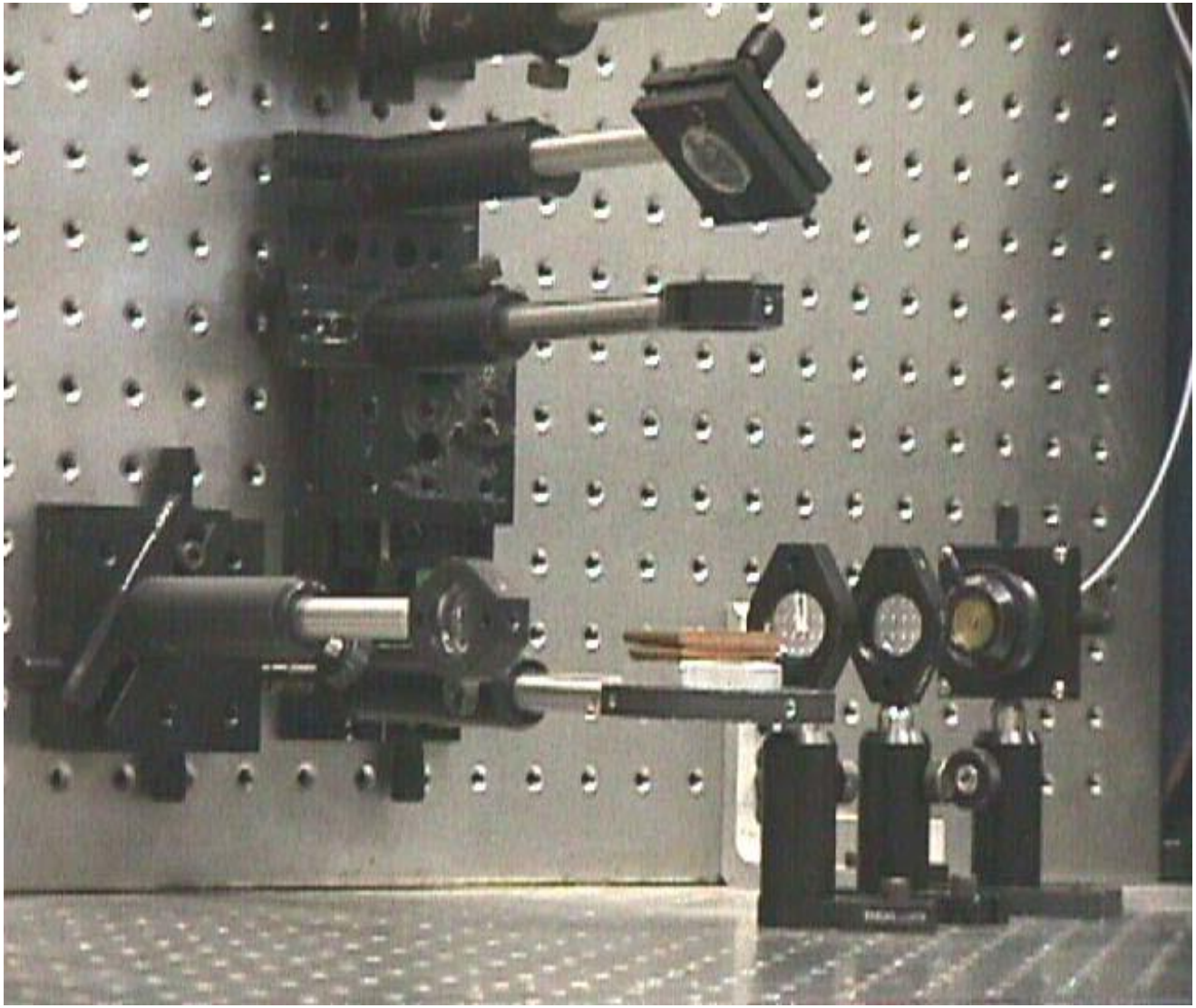
Pb S/B ~2.5

One μs ΔT

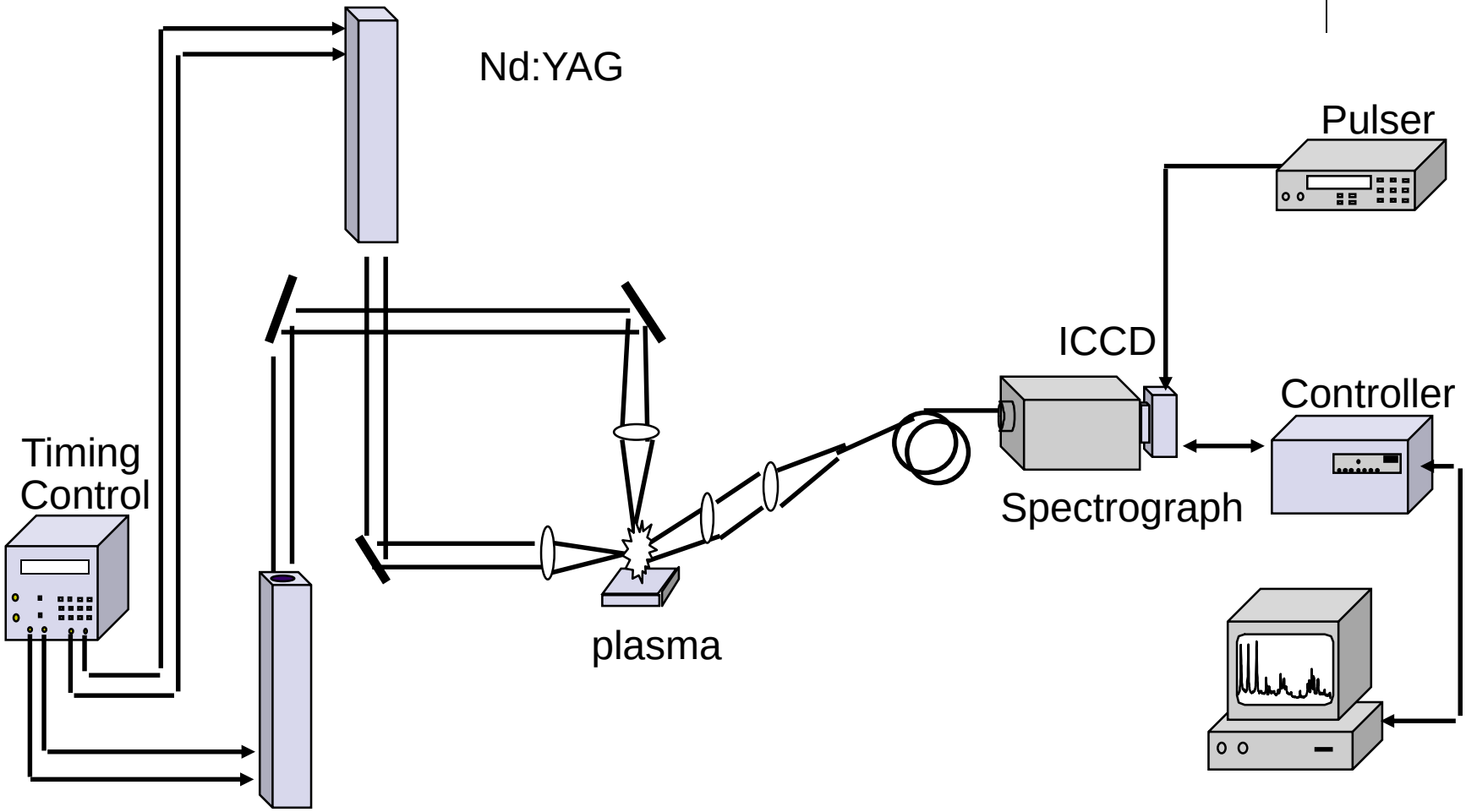


Pb S/B ~ 6

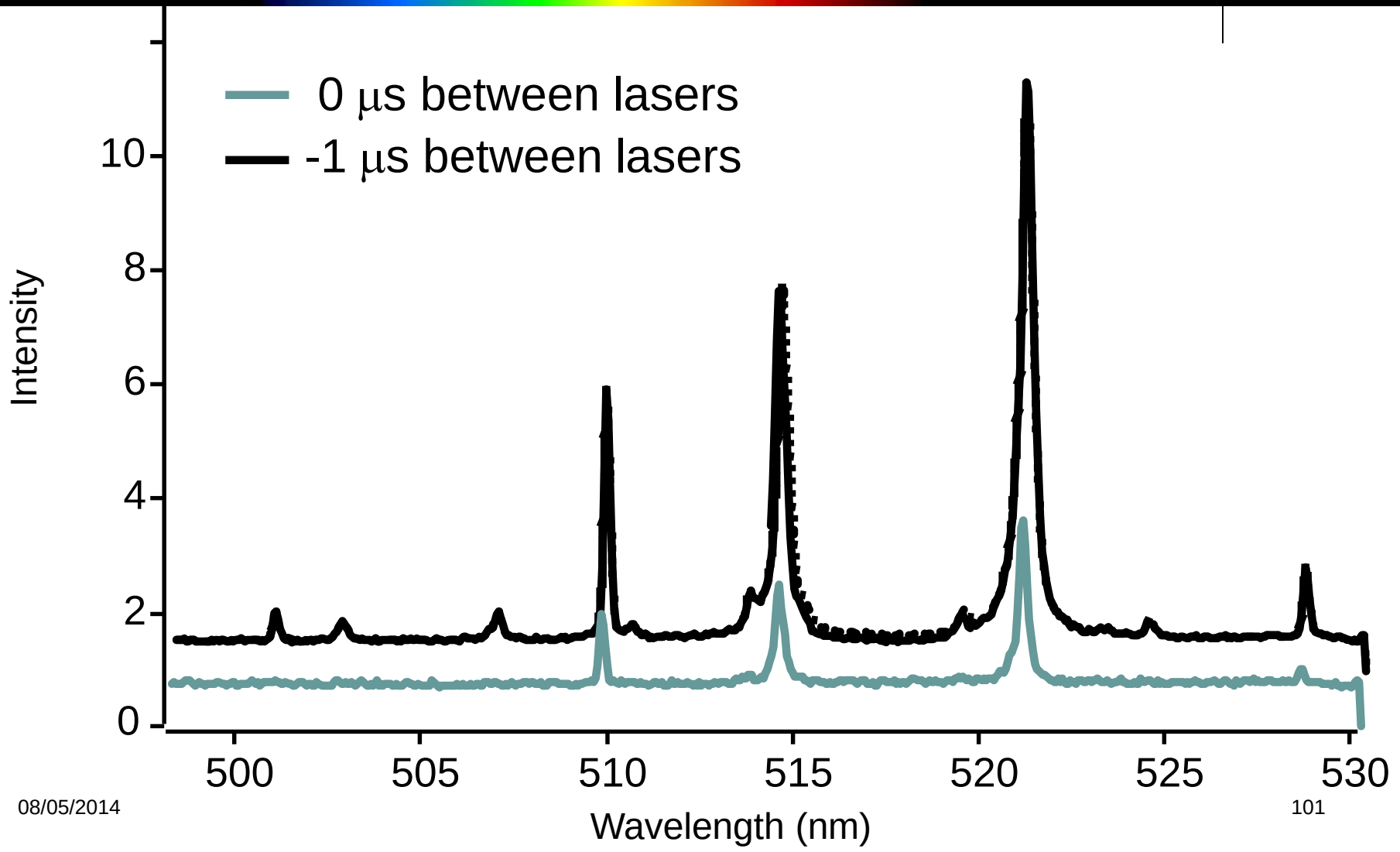
Orthogonal Dual-Pulse LIBS



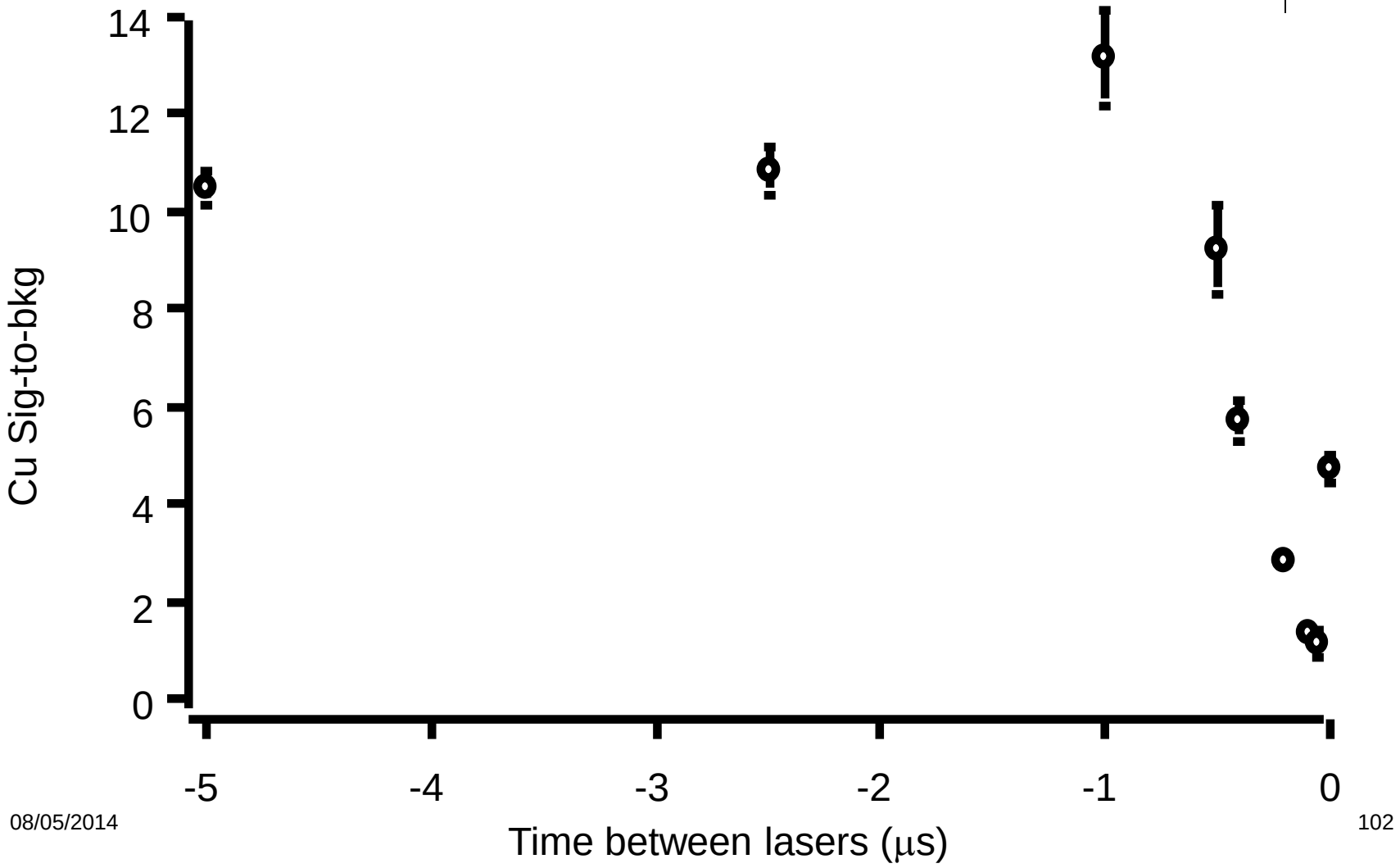
Orthogonal Dual-Pulse LIBS



Orthogonal Dual-Pulse LIBS Enhancement for Cu

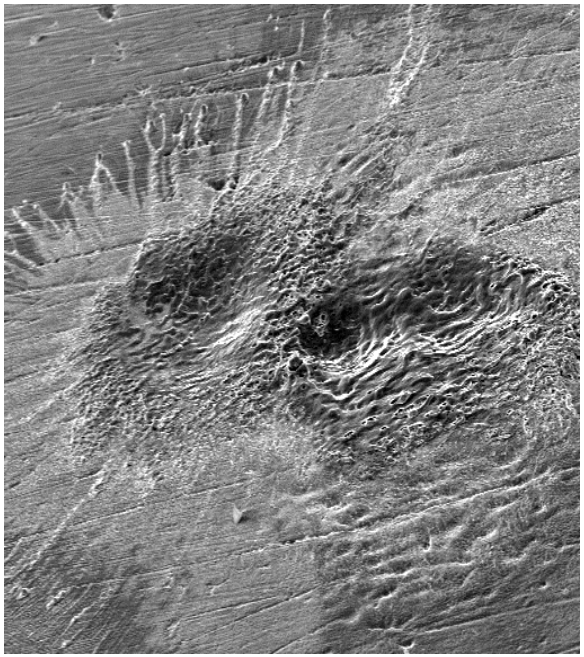


Enhancement of Copper Emission Using Non-Ablating Prespark



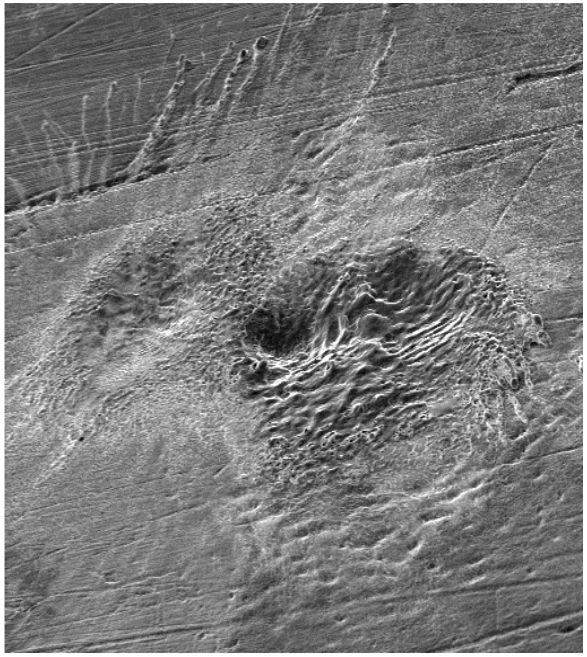
Orthogonal Dual-Pulse LIBS Geometry SEM Craters for Copper

0 μs ΔT



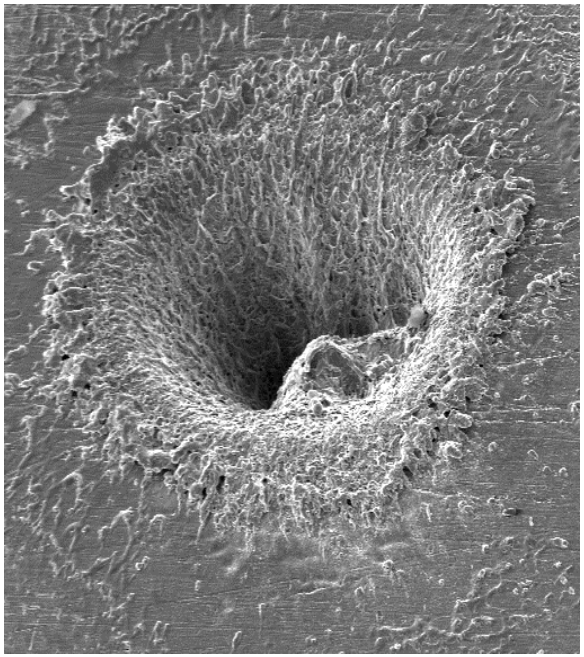
150 μm

1 μs ΔT



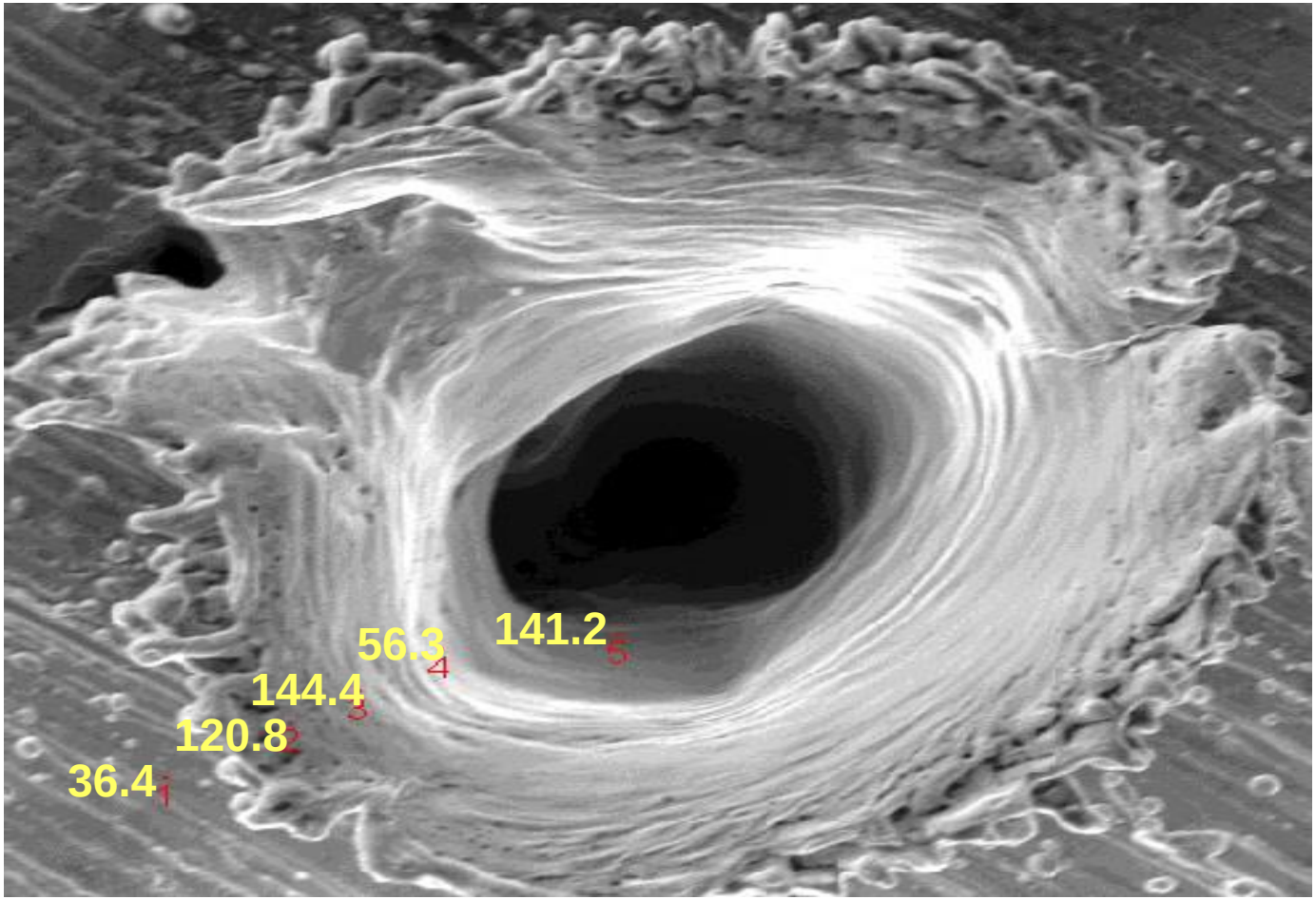
150 μm

20 μs ΔT



176 μm

2.86% Zinc at Low Power



NATIONAL INSTITUTE OF LASER ENHANCED SCIENCES

N . I . L . E . S

جامعة القاهرة

Creation of Ultrafast Ultra-Broadband High Energy Laser Pulses

Prof. Walid Tawfik

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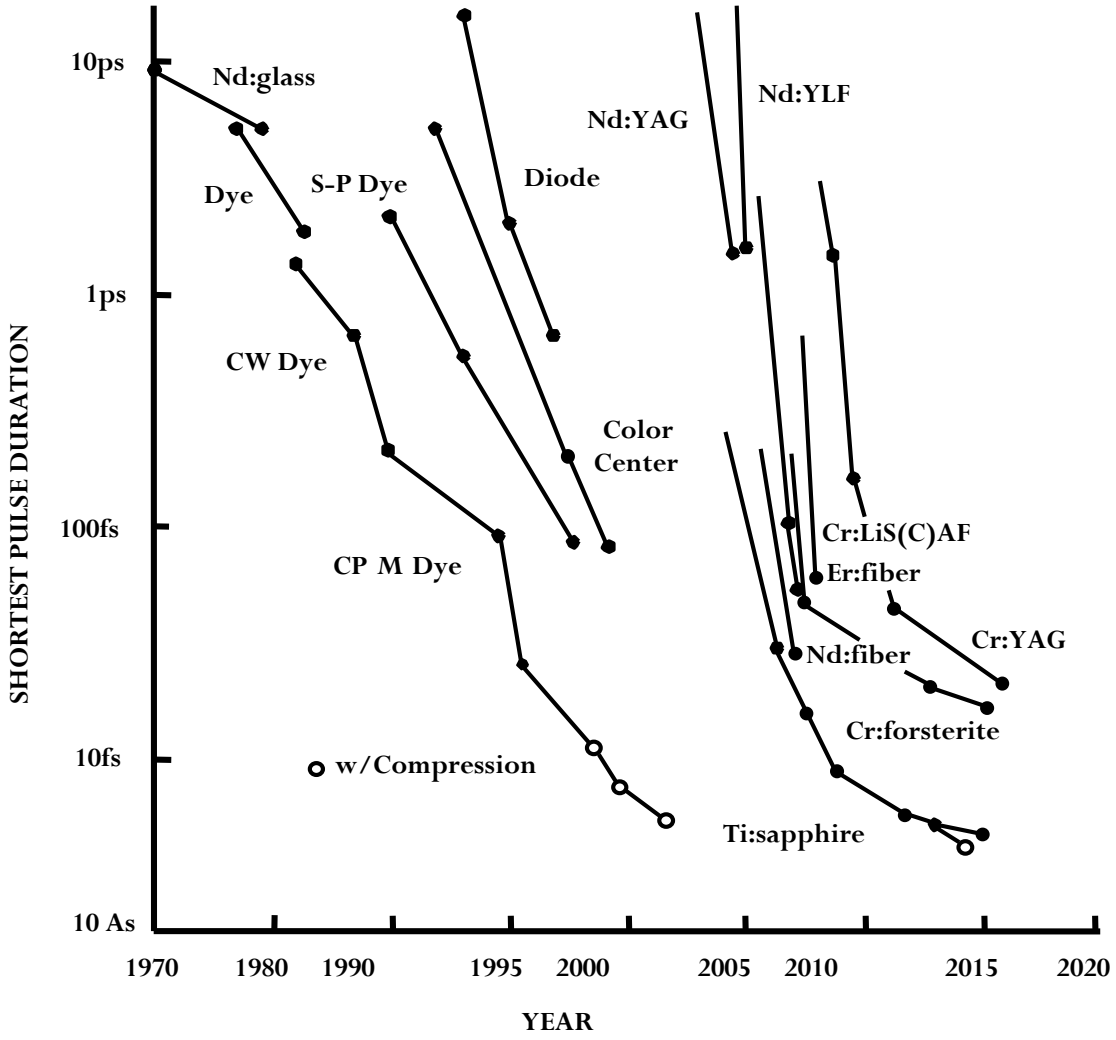
Outlines

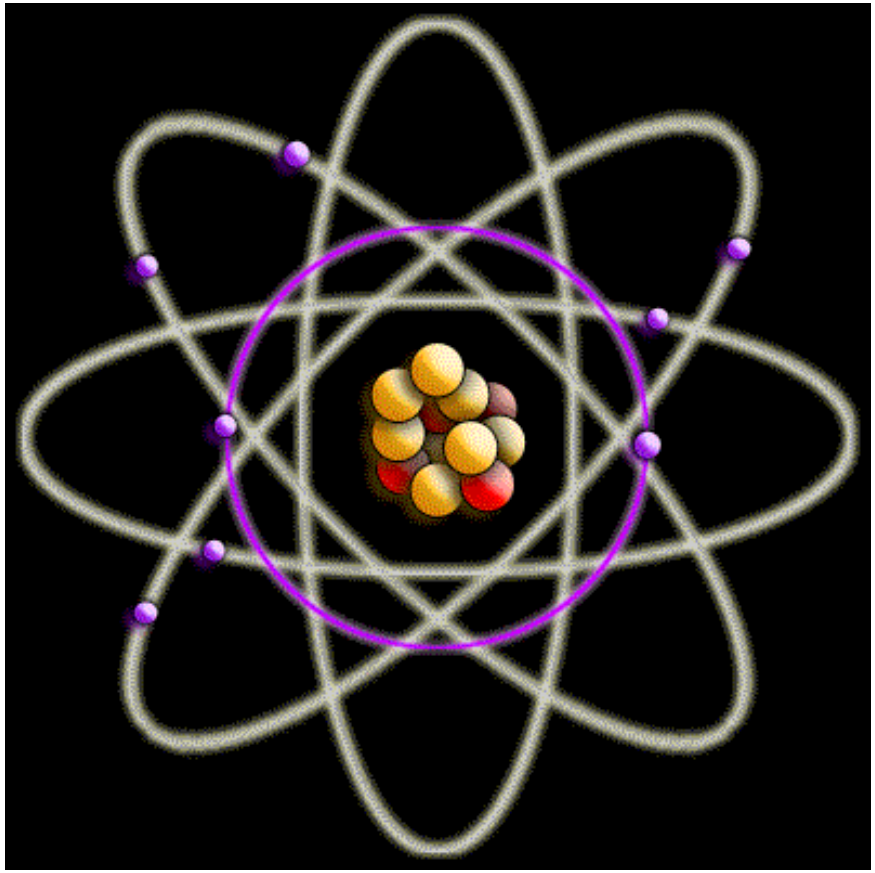
- Why we need ultrafast white light (tunable) high-power laser pulse?
- Generation and measurements of ultrafast laser pulses with via a one meter neon-filled hollow-fiber.
- Experimental setup layout and Output characteristics using SPIDER.
- Controlling the Tunable broad-bandwidth ultrafast laser pulses.
- Conclusion.

The ultrafast progress has been amazing!



The shortest pulse vs. year (for different media)

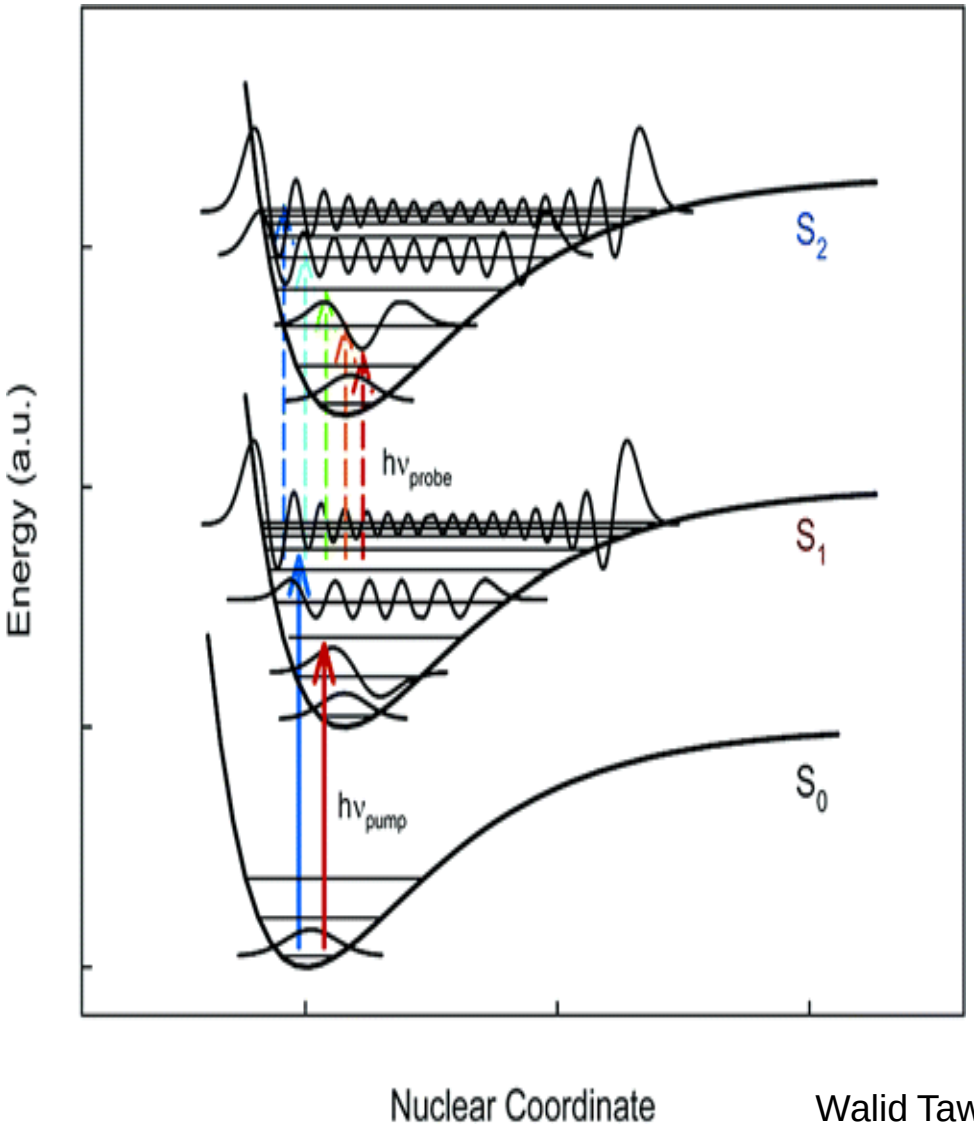
Why try to make ultrafast pulses?



152 attoseconds

Molecular vibrations can also be very fast.

Transient Absorption – in complex System



- Vibrational Relaxation (VR), Intersystem Crossing (ISC), and Internal Conversion (IC)
- Aspects of VR
  - Pump wavelength dependence
    - Density of states
  - Probe wavelength dependence
  - Franck-Condon Factors
- Full-spectrum, Kinetic trace
- Needed Information
  - Steady State absorption and emission
  - geometry
  - Electron configuration

What's so Special About Femtosecond Lasers???

- *Short optical pulse.*
  - Most of energy dissipation **(heat dissipation)** and transfer processes occur on the time scale larger than **100 fs.**
  - Femtosecond laser pulses enable one to excite the species studied "instantly" ($t_{exc} << t_{rel}$)
  - Dynamics of the excited state can be monitored with high temporal resolution (~ *0.5* $\tau_{pulse} \approx$ *12-50 fs for most of commercial lasers*)
  - *Visualization of ultrafast dynamical processes (fluorescence, excited state absorption)*
- *High peak power of the light*
  - $I \sim J/\tau_{pulse}$, *I* – Power, *J* – *pulse energy.*
  - 1 mJ pulse with 10 ns duration - **0.1 MW**
  - 1 mJ pulse with 100 fs duration - **10 GW**
  - *Non-linear spectroscopy and materials processing (e.g., multi-photon absorption, optical harmonics generation, materials ablation, etc.)*

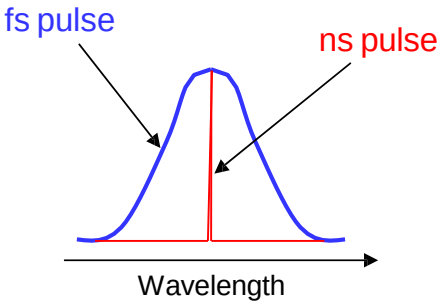
W. Kaiser, ed., "*Ultrashort Laser Pulses: Generation and Applications",* Springer-Verlag, Berlin, **1993**

How to Prepare a Femtosecond Pulse I

Femtosecond laser pulses are usually ***Fourier transform-limited pulses***

$\Delta\omega\Delta t \approx 2\pi$ ➡ $\Delta\omega \approx 2\pi/\Delta t$ ➡ Large spectral bandwidth for short pulses

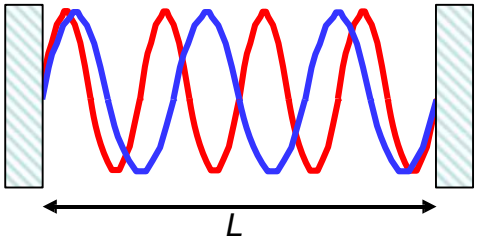
$\Delta\lambda \approx \lambda^2/(c\,\Delta t)$ $\Delta\lambda \approx$ 21 nm for 100 fs pulses with $\lambda_0 = 800$ nm



Large bandwidth limits the choice of the laser active medium (broad-band materials only, e.g., Ti:Sapphire, laser dyes) and laser cavity design (no bandwidth limiting elements, such as narrowband mirrors)

How to Prepare a Femtosecond Pulse II

Laser mode – combination of frequency (ω) and direction (***k***) of the electromagnetic wave allowed by the laser cavity geometry.

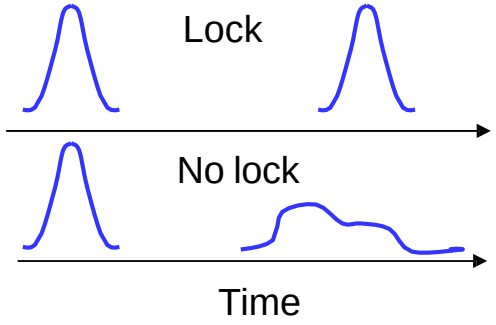


The spectrum of laser modes is not continuous $\lambda_n = 2L/n$

$$I(t) = \sum_{n=0}^{N} A_n \sin(\omega_n t + \varphi_n)$$

Laser pulse as a sum of modes

Relative phase of the modes has to be constant (locked) in order to obtain a stable output pulse



Passive Mode-Locking

Saturable absorption is a property of materials where the absorption of light decreases with increasing light intensity. Most materials show some saturable absorption, but often only at very high optical intensities (close to the optical damage). At sufficiently high incident light intensity, atoms in the ground state of a saturable absorber material become excited into an upper energy state at such a rate that there is insufficient time for them to decay back to the ground state before the ground state becomes depleted, and the absorption subsequently saturates.

Saturable absorption examples:

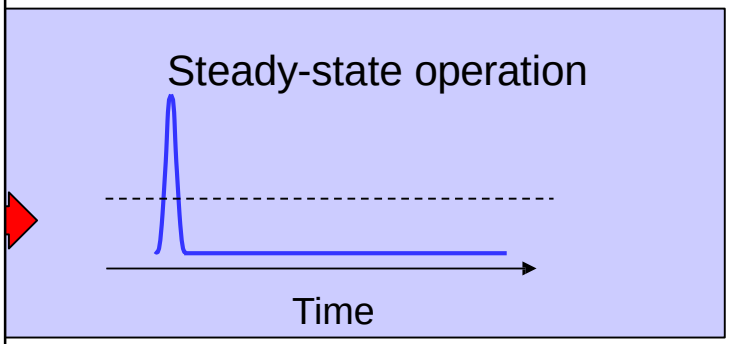
*Gallium arsenide (GaAs)

*Thin layers of carbon nanotubes (CNT)

*single or multiple graphene layers

*$Co^{2+}:MgAl_2O_4$

* $Cr^{2+}:ZnS$ and $Cr^{2+}:ZnSe$

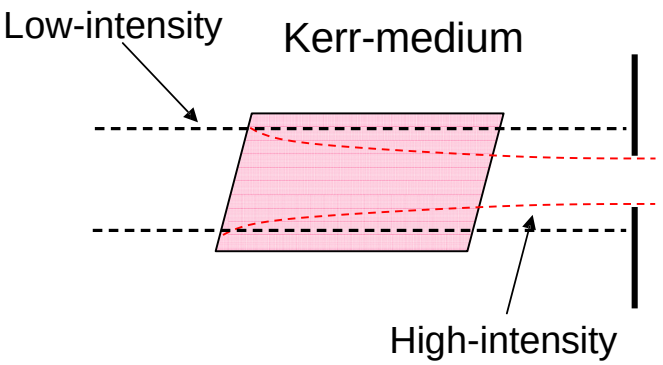


Passive Mode-Locking II

One Cycle of light pulse 800 nm is
T = λ / c
= 800 nm / 3 x 10^8
= **2.6 fs**

Kerr-lens mode-locking

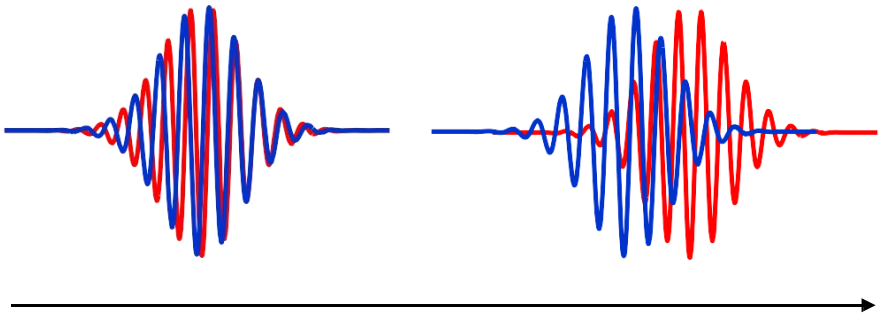
- Kerr's effect – inetnsity-dependent index of refraction: $n = n_0 + n_2 I$
- The e/m field inside the laser cavity has Gaussian distribution of intensity which creates similar distribution of the refractive index.
- High-intensity beam is self-focused by the photoinduced lens.



- High-intensity modes have smaller cross-section and are less lossy. Thus, Kerr-lens is similar to saturating absorber!
- Some lasing materials (e.g. Ti:Sapphire) can act as Kerr-media
- Kerr's effect is much faster than saturating absorber allowing one generate very short pulses (~5 fs).

Group Velocity Dispersion (GVD)

Optical pulse in a transparent medium stretches because of GVD



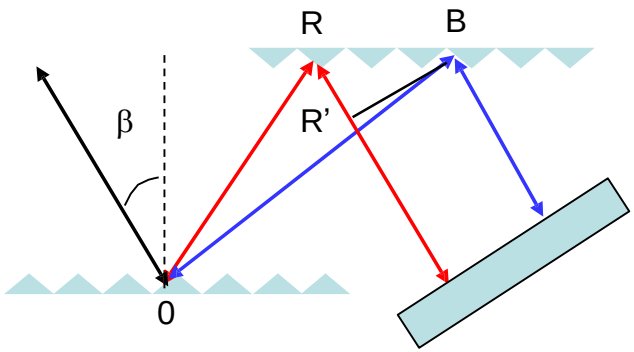
•v = c / n – speed of light in a medium
• n –depends on wavelength, $dn/d\lambda < 0$ – normal dispersion

•Because of GVD, red components (longer wavelengths) of the pulse propagate faster than blue components (shorter wavelengths) leading to pulse stretching (aka “chirp”).
• Uncompensated GVD makes fs laser operation impossible
• GVD can be compensated by material with *abnormal* dispersion

GVD Compensation

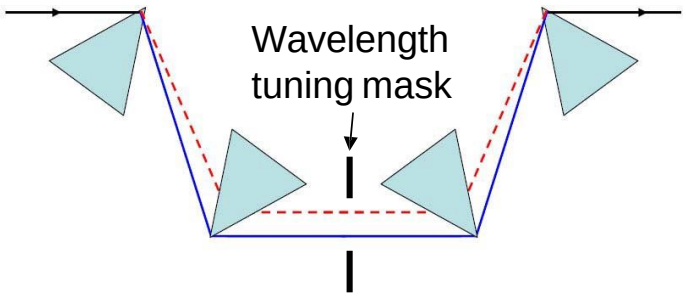
GVD can be compensated if optical pathlength is different for “blue” and “red” components of the pulse.

Diffraction grating compensator



If OR + RR’ > OB, GVD < 0

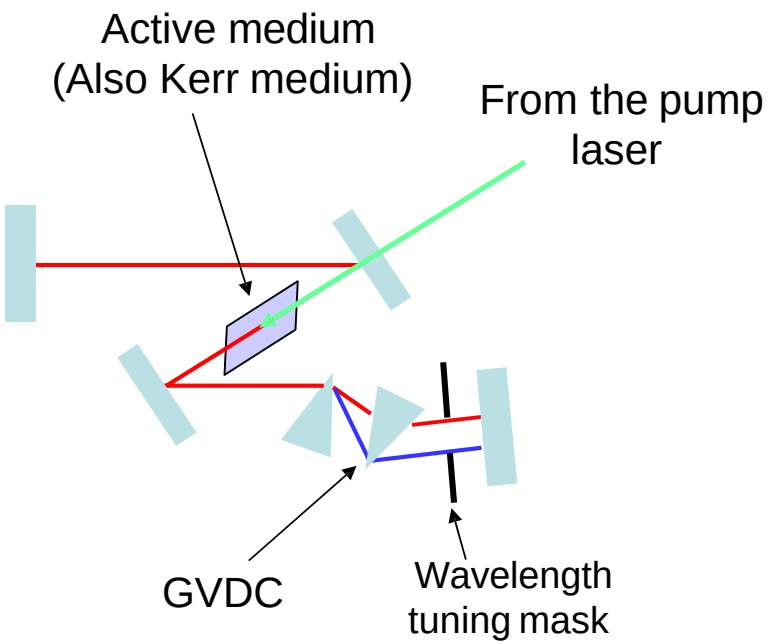
Prism compensator



“Red” component of the pulse propagates in glass more than the “blue” one and has longer optical path (n x L).

Typical fs Oscillator

Typical Ti:Sapphire fs Oscillator Layout



- Tuning range 690-1050 nm
- Pulse duration > 5 fs (typically 50 -100 fs)
- Pulse energy < 10 nJ
- Repetition rate 40 – 1000 MHz (determined by the cavity length)
- Pump source:
  Ar-ion laser (488+514 nm)
  DPSS CW YAG laser (532 nm)
- Typical applications:
  time-resolved emission studies, multi-photon absorption spectroscopy and imaging

O. Zvelto, “Principles of lasers”, Plenum, NY (2004)

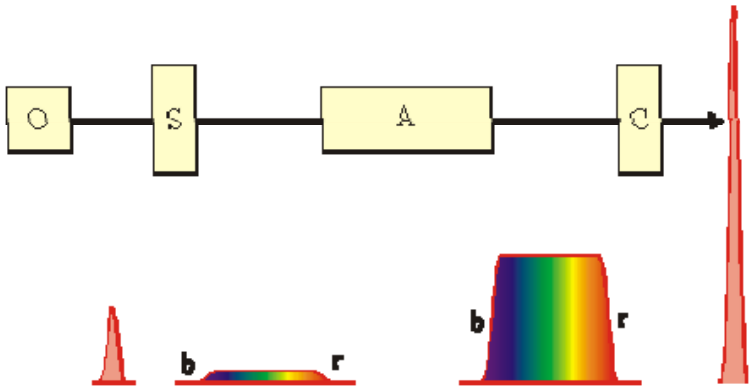
Amplification of fs Pulses

Due to high intensity, fs pulses can not be amplified as is.

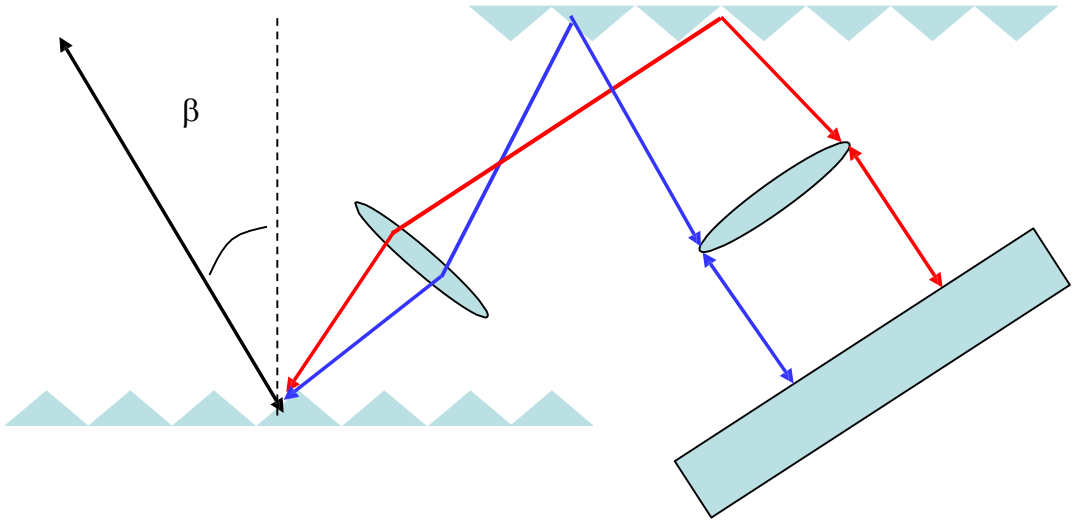
Recipe for the amplification:

Chirped pulse amplifier (CPA)

- Stretch the pulse in time, thus reducing the peak power ($I = J / t_{pulse}$!) (typically the pulse is stretched up to hundreds of ps)
- Amplify the stretched pulse
- Compress the pulse

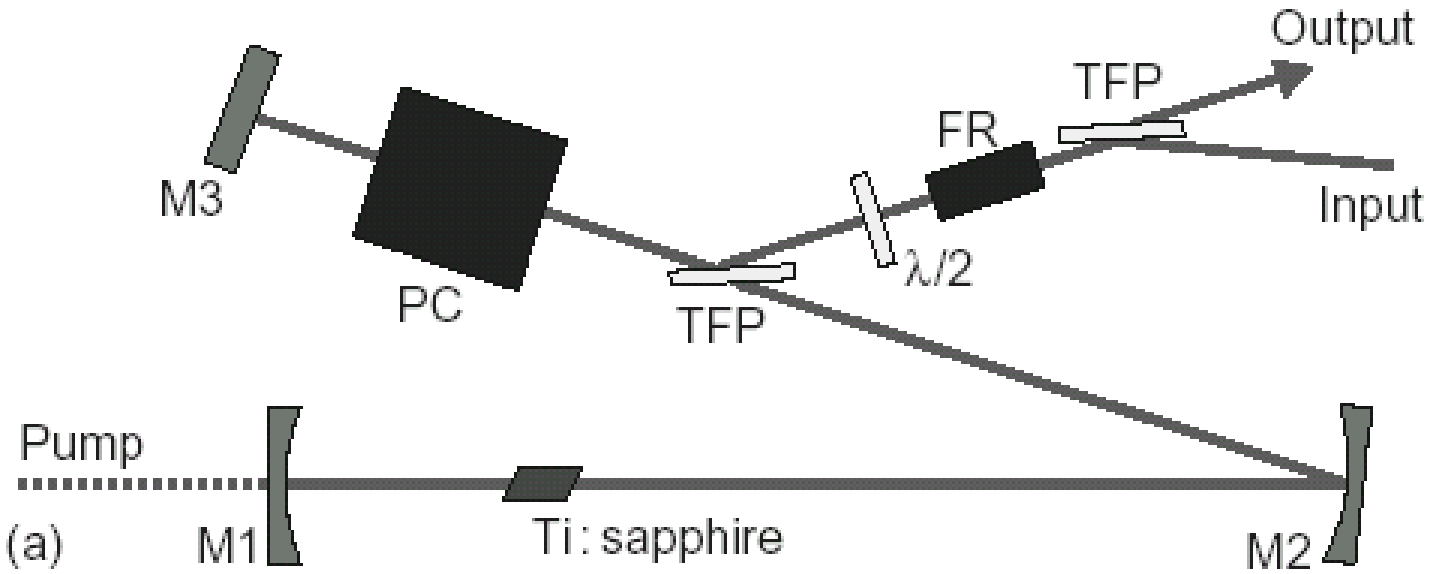


Pulse Stretcher



•Pulse stretcher utilizes the same principle as compressor: separation of spectral components and manipulation with their delays
•Compressor can converted into stretcher by addition of focusing optics "flipping" paths of red and blue components.

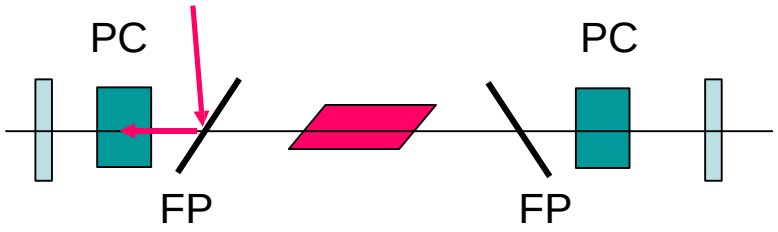
Regenerative amplifier



scheme for regenerative amplification. The design is often at a 10–20-Hz repetition rate. The Ti : sapphire rod is usually ca. 20 mm long and doped for 90% absorption. TFP, thin-film polarizing beamsplitter; PC, Pockels cell; FR, Faraday rotator; $\lambda/2$, half-wave plate. In (a), M1 is 150 mm radius of curvature, M2 is 1 m and M3 is flat. In (b) M1 is −20 m and M2 is +10 m.

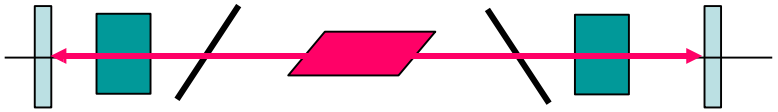
Regenerative Amplifier

Cavity dumping

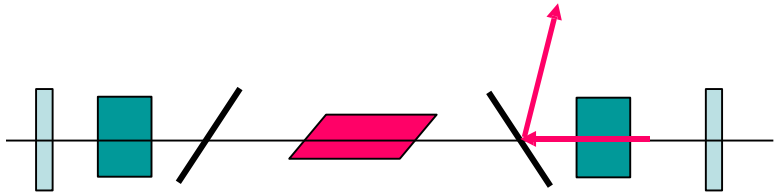


Injection of the pulse from stretcher (FP – film polarizer, PC – Pockels cell), Pockels cell rotates polarization of the seeding pulse

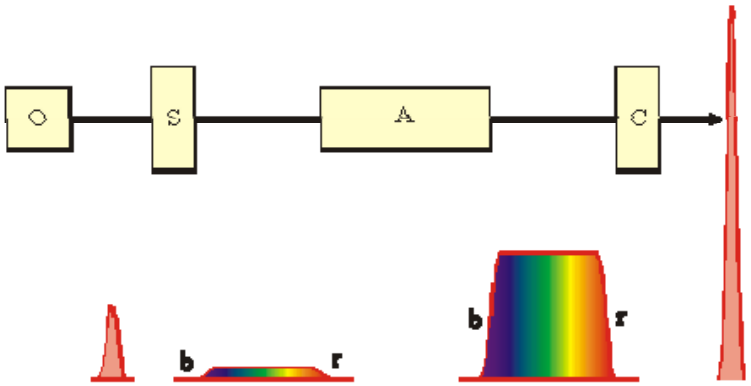
Amplification



Ejection of the pulse into compressor



Typical CPA

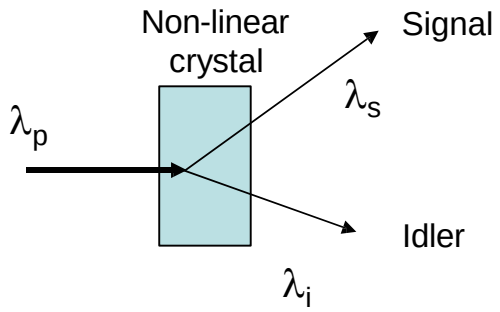


- Repetition rate ~ 1 KHz
- Pulse duration 50-150 fs
- Pulse energy 1 mJ
- Wavelength – usually fixed close to 800nm
- Typical applications: pumping optical frequency converters, non-linear spectroscopy, materials processing

Frequency Conversion of fs Pulses

With fs pulses non-linear optical processes are very efficient due to high intensity of input light: $I_{out} = A\ I_{in}^{m}$

Parametric down-conversion



$$1/\lambda_p = 1/\lambda_s + 1/\lambda_i$$
$$\boldsymbol{k}_p = \boldsymbol{k}_s + \boldsymbol{k}_i$$

Pump : 800 nm, 1mJ, 100 fs
Signal: 1100 -1600 nm, 0.12 mJ
Idler: 1600 – 3000 nm, 0.08 mJ

Optical harmonic generation

Second harmonic

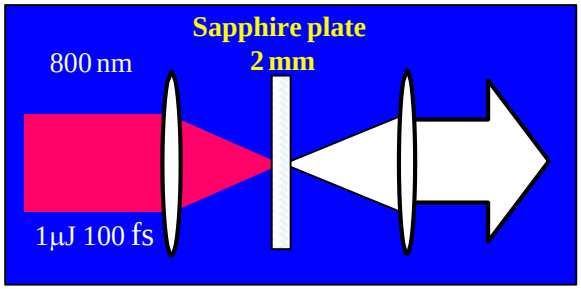
$$1/\lambda_{SH} = 2/\lambda_F$$
$$\boldsymbol{k}_{SH} = 2\,\boldsymbol{k}_F$$

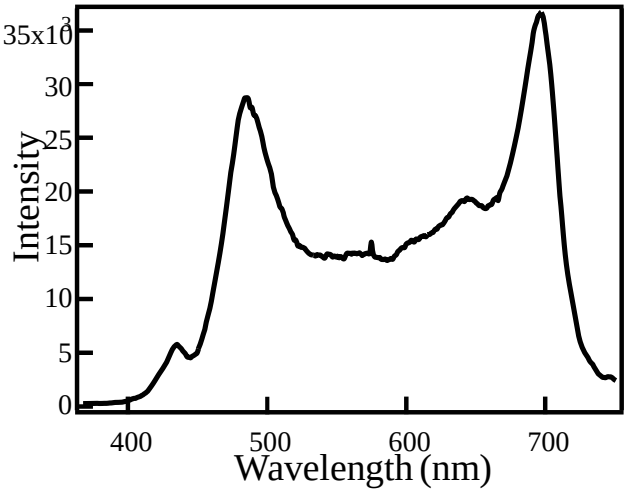
Pump : 800 nm, 1mJ, 100 fs
SHG: 400 nm, 0.2 mJ

Harmonic generation can be used to upconvert signal or idler into the visible range of spectrum

Femtosecond Continuum

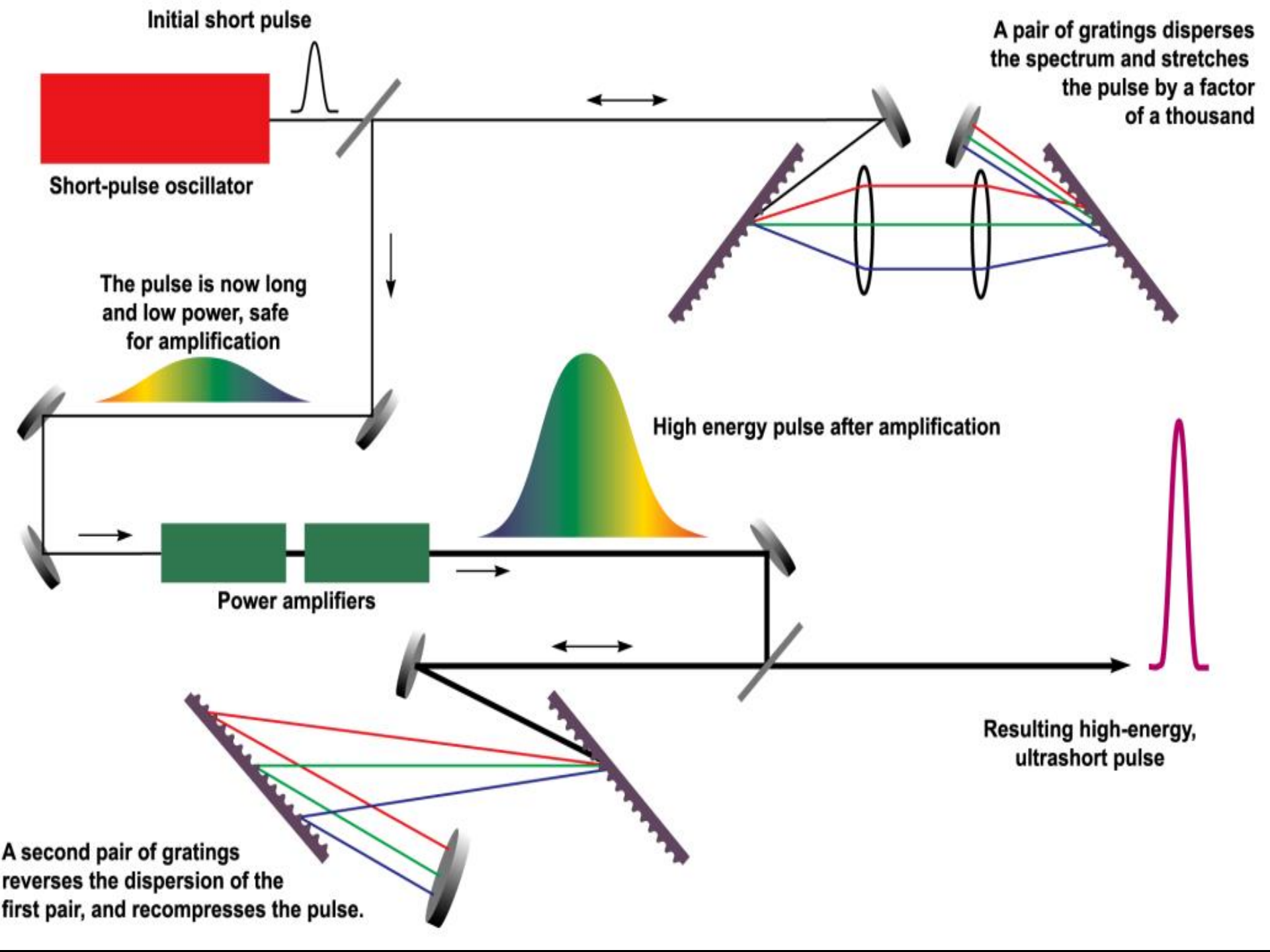
White-light continuum generation





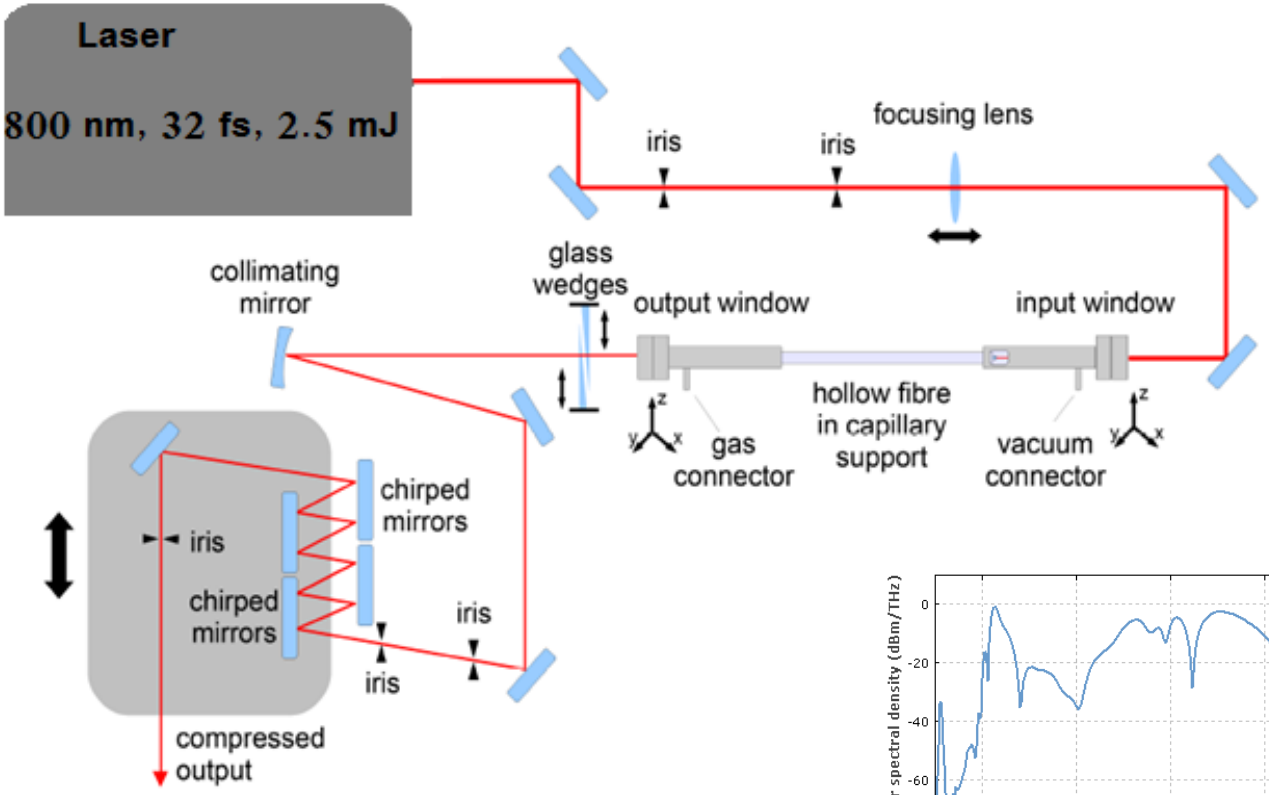
- Self-focusing and self-phase-modulation broadens the spectrum
- Extremely broad-band, ultrafast pulses (Vis and IR ranges)
- Strongly chirped

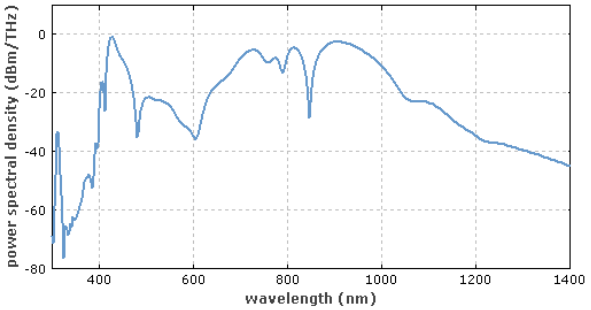
1.R. L. Fork et al, **8** *Opt.Lett., p. 1,* (1983)


Initial short pulse
Short-pulse oscillator
A pair of gratings disperses the spectrum and stretches the pulse by a factor of a thousand
The pulse is now long and low power, safe for amplification
High energy pulse after amplification
Power amplifiers
Resulting high-energy, ultrashort pulse
A second pair of gratings reverses the dispersion of the first pair, and recompresses the pulse.

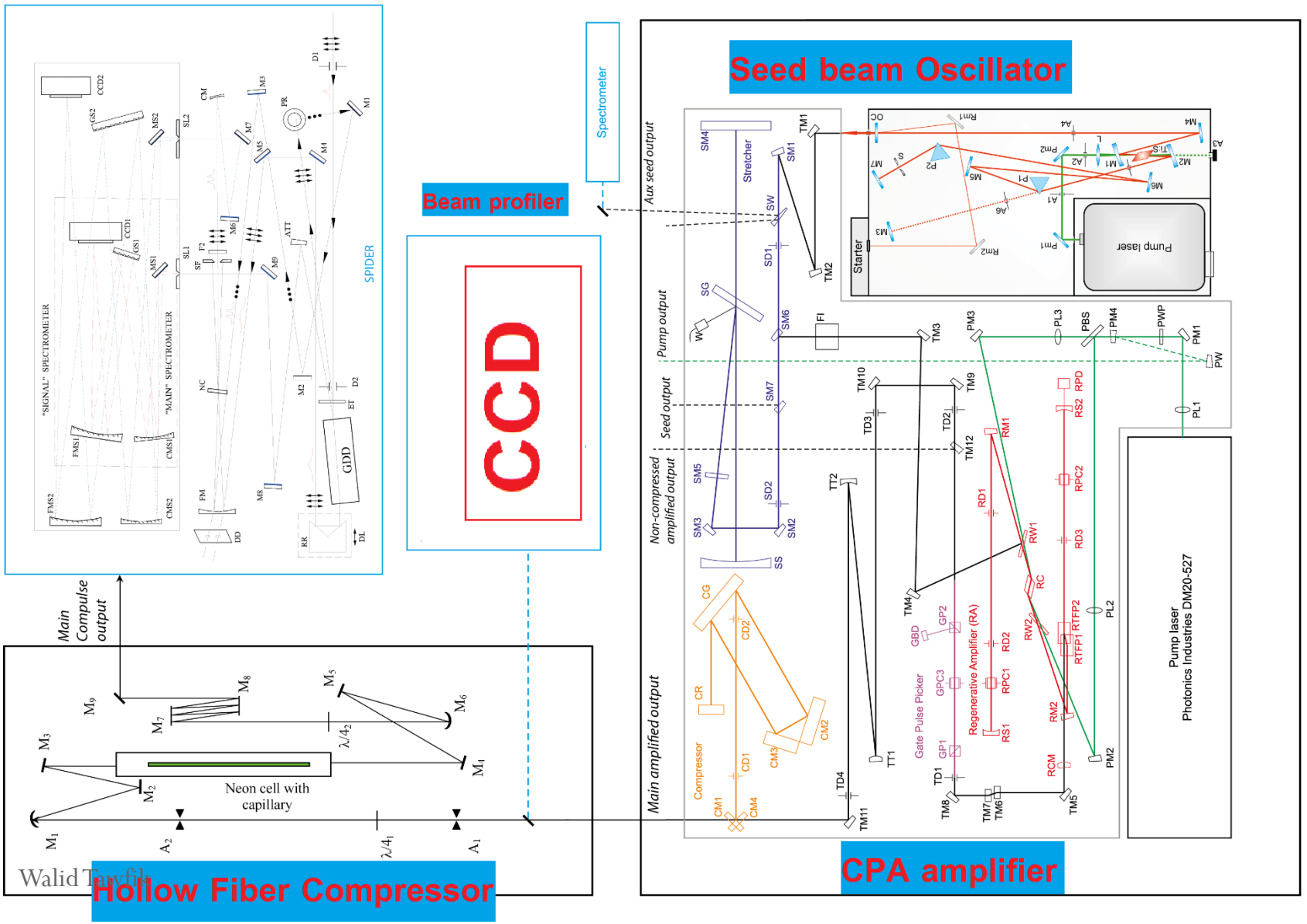
Schematic diagram of the few-cycle white light generator.

The experimental setup for the project, where the laser system provides 32-fs pulses and energies of up to 2.5 mJ at a repetition rate of 1 kHz. The amplified pulses will be focused into a 1-m-long differentially pumped hollow core fiber (inner core diameter of 250 μm). The spectrally broadened pulses at the output of the fiber system will be compressed by 10 bounces from double-angle technology CMs . A pair of fused silica wedges will be used to fine tune the pulse compression.

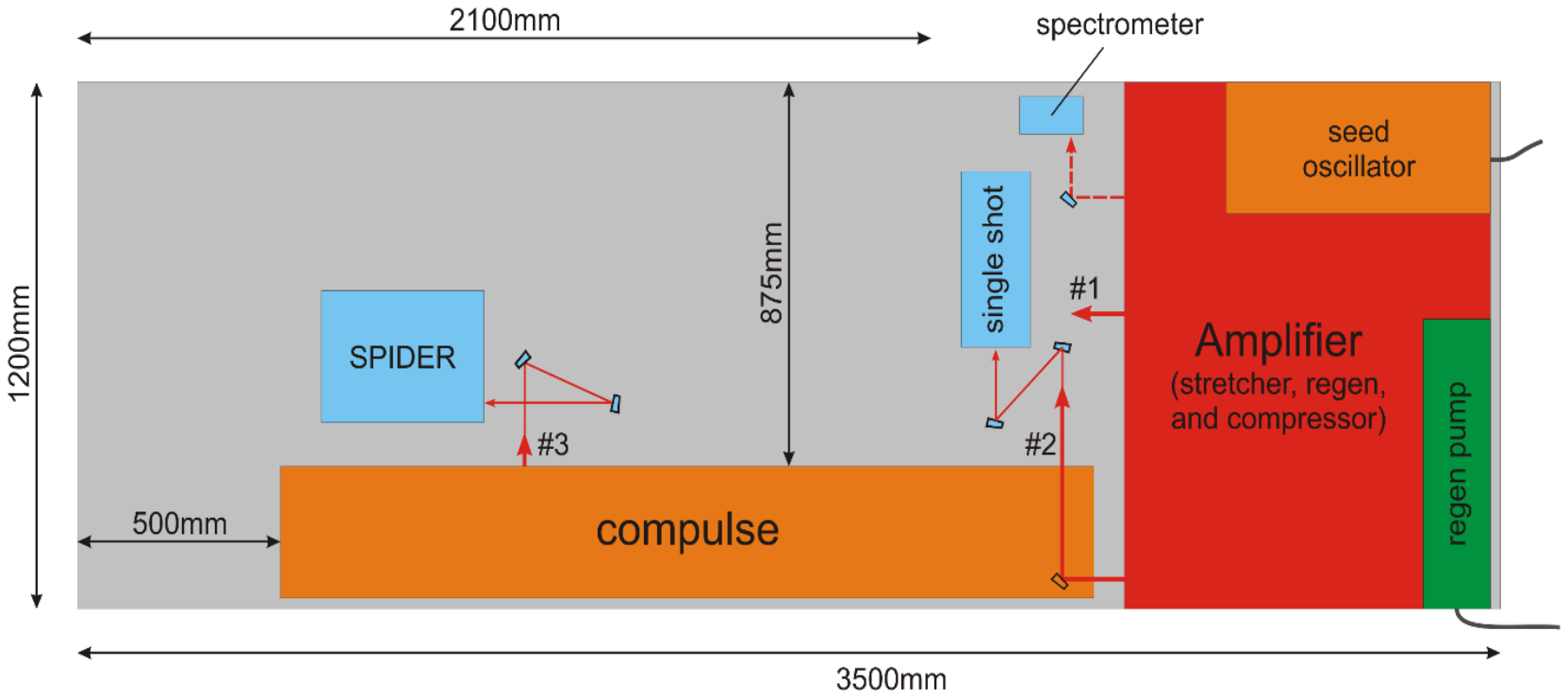




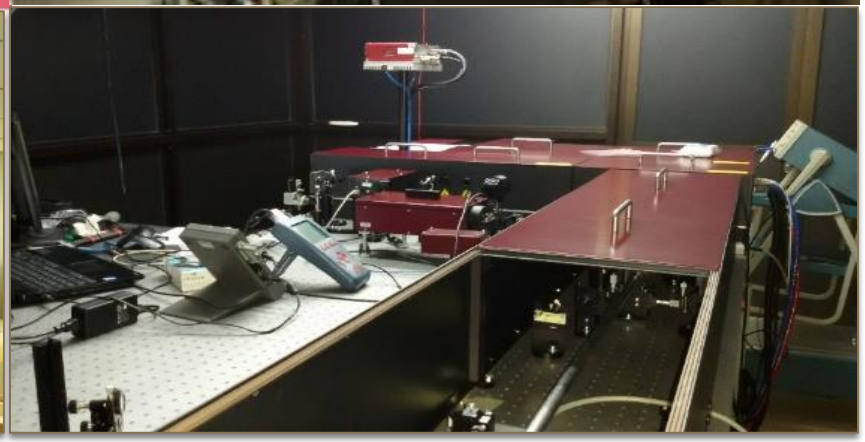
Optical layout of the few-cycle white light generator.



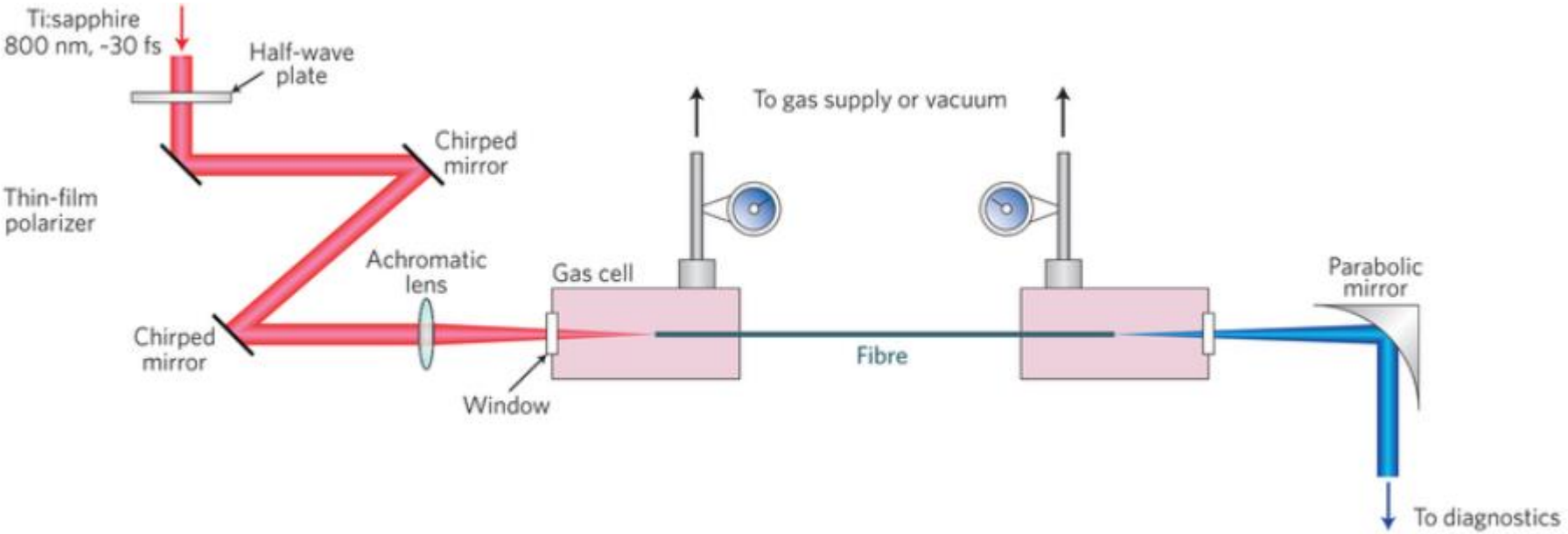
Optical layout of the table-top ultrafast laser

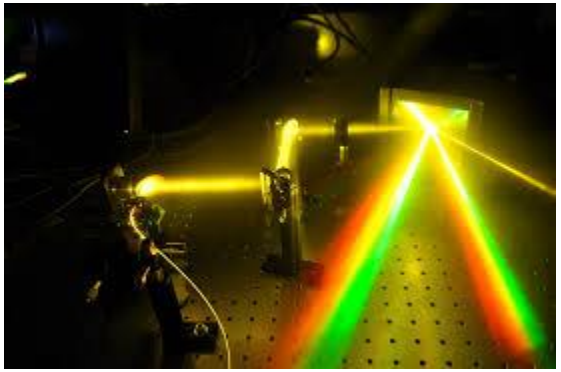


The homemade semi-clean room and the table-top system



Optical layout of the Hollow-fiber compressor.





[Poster 42]

Attosecond light facility construction in CASTECH

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ATTO3 conference Japan July 6-8 2011

Since generation of isolated attosecond pulse by few cycle laser, which pulse duration is shorter than 5fs, Attosecond pulse is a unique tool to observe ultrafast phenomena: time scale of reaction is shorter than 1fs. The authors will present progress of Attosecond light facility construction in CASTECH. Fig 1. Shows pictures of AS light facility in CASTECH.

Fig. 1 3D modeling & Pictures

Paul Corkum

Dr. Walid Tawfik

Dr. Walid Tawfik

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Generation of Few Cycle Femtosecond Pulses via Supercontinuum in a Gas-Filled Hollow-Core Fiber

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[2]*National Institute of laser Enhanced Sciences, Cairo University, Cairo,*
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[2]*Egypt*

1. Introduction

The interaction of intense short nano- and picosecond laser pulses with plasma leads to reach variety of important applications, including time-resolved laser induced breakdown spectroscopy LIBS (Walid Tawfik et al., 2007), soft x-ray lasers (X. F. Li et al., 1989), and laser-driven accelerators (W. P. Leemans et al., 1989). In most cases the useful output - whether it be photons or accelerated particles - increases with the distance over which the laser-plasma interaction occurs, at least up to some upper limit which might, for example, be set by the onset of phase mismatching. The strength of any such interaction will depend on


Squeezing the light pulse

White light generation by Self Phase Modulation

$n(I) = n_0 + n_2 I + \ldots$

The electrical laser field is

$E(x,t) = E(t)\cos(\omega t - kx)$

$\varphi = \omega t - kx = \omega t - \omega n x/c$
$= \omega(t - n_0 z/c) - n_2\, \omega z/c I(t)$

$\omega = d\varphi/dt = \omega - A\, dI/dt$

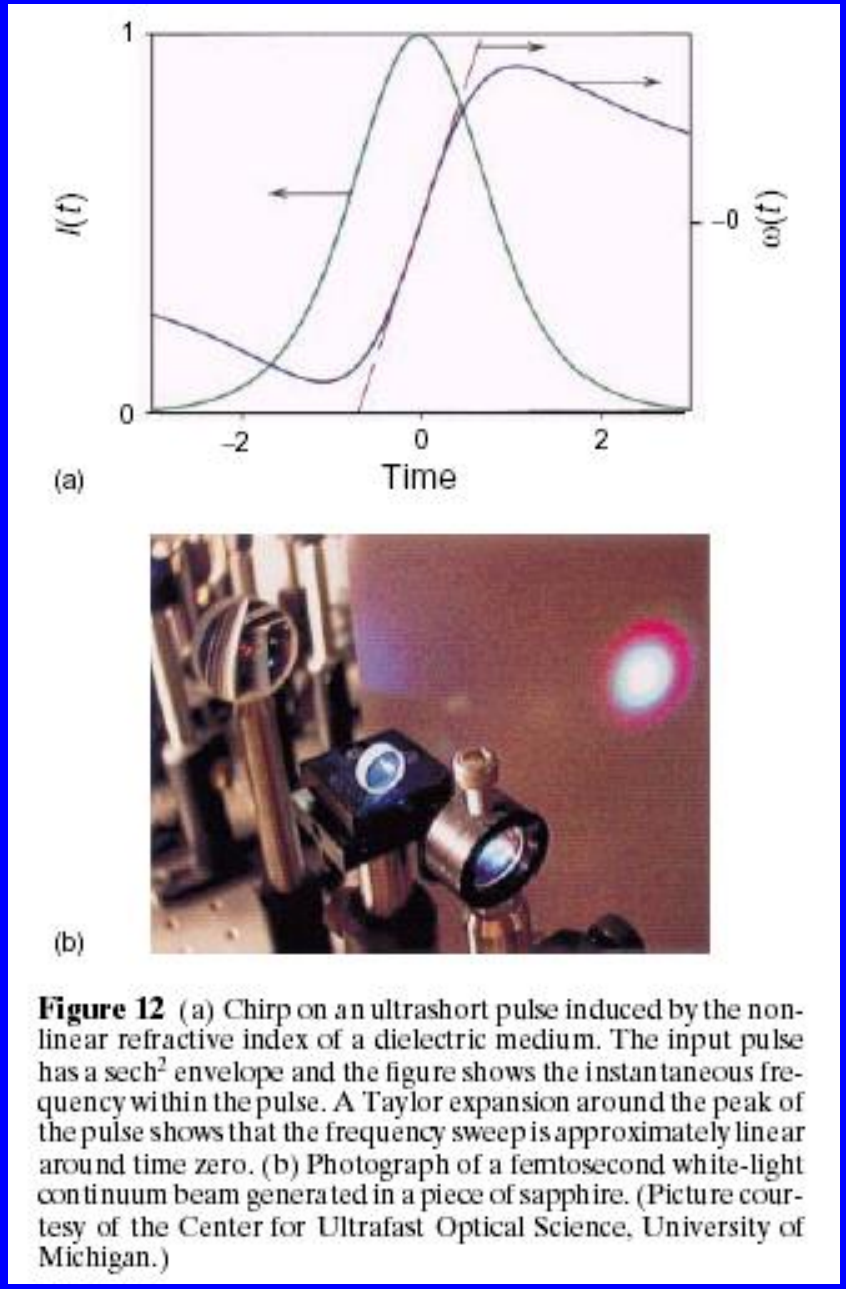
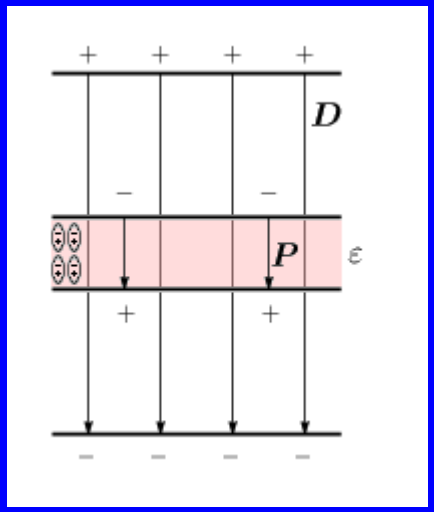


Figure 12 (a) Chirp on an ultrashort pulse induced by the nonlinear refractive index of a dielectric medium. The input pulse has a sech^2 envelope and the figure shows the instantaneous frequency within the pulse. A Taylor expansion around the peak of the pulse shows that the frequency sweep is approximately linear around time zero. (b) Photograph of a femtosecond white-light continuum beam generated in a piece of sapphire. (Picture courtesy of the Center for Ultrafast Optical Science, University of Michigan.)



Light-matter interaction

Electric field make dielectric polarization

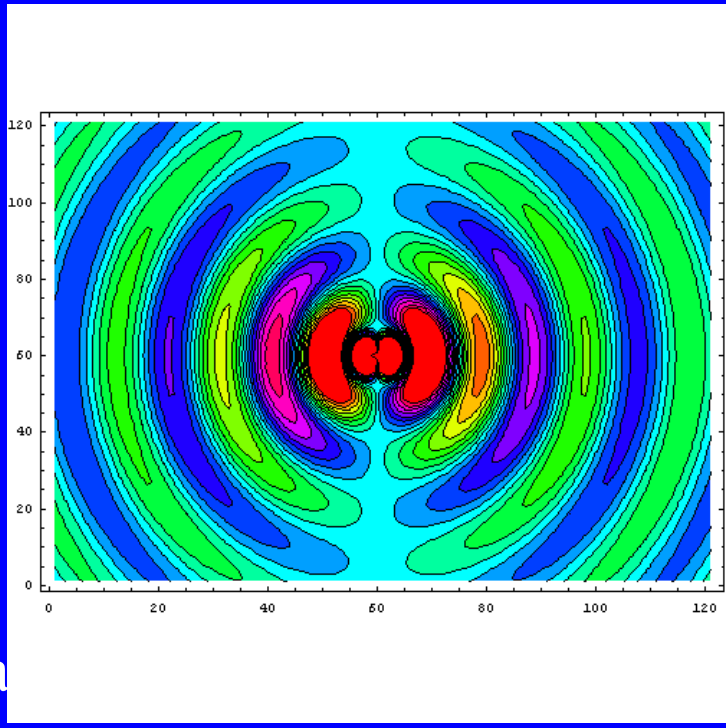
$$P = \varepsilon_0(\chi^{(1)}E + \chi^{(2)}EE + \chi^{(3)}EEE\cdots)$$

Emission from dipole

oscillating in vertical direction

E : exp(-iωt)

E*E : exp(-i2ωt)



Second harmonic generation (SHG)

using **a *non-linear* crystal**

within some limitation from physica

Pulse Self-Phase Modulation SPM

- The influence of an applied field that causes also the induced polarization P from the electric dipoles to be nonlinear with the electric field E, and follow the nonlinear relation (Shen et al., 1984)
-

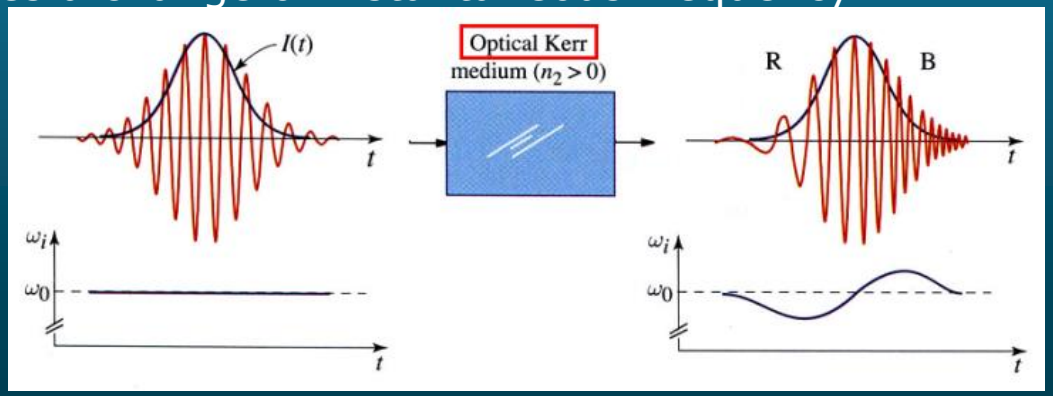
$$P = \varepsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + ...]$$

$x^{(n)}$, *n*th-order nonlinear susceptibilities of the medium, respectively.

Self-Phase Modulation **(SPM)** is caused by nonlinear **Kerr effect** , **$n(I)=n+n_2 I$** , and a phase **$\Delta\varphi = -n_2 I k_0 z$** induced by wave traveling a distance z in Kerr medium with time varying intensity I(t)

$$\Delta\varphi(t) = -n_2 I(t) k_0 z \quad \xrightarrow{d\varphi/dt} \quad \Delta\omega_i = -n_2 \frac{dI}{dt} k_0 z$$

this gives a change of instantaneous frequency



Supercontinuum Generation in Hollow Fibers

- The radial intensity profile of EH_{1m} modes is given by $I_c(r) = I_{c0}\, J_0^2\,(\upsilon_m\ r/a\,)$ where J_0 is the zero-order Bessel function, a is the capillary radius, υ_m is the m th zero of $J_0(r)$, I_{c0} is the peak intensity. The complex propagation constant $\beta(\omega)$ of the EH_{1m} mode is given by (Shen et al., 1984)

$$\beta(\omega) = \frac{\omega\eta(\omega)}{c}\left[1 - \frac{1}{2}\left(\frac{\upsilon_m c}{\omega\eta(\omega)a}\right)^2\right] + \frac{i}{a^3}\left(\frac{\upsilon_m c}{\omega\eta(\omega)}\right)^2 \frac{\upsilon^2(\omega)+1}{\sqrt{\upsilon^2(\omega)-1}} \qquad (7)$$

- where $\eta(\omega)$ is the refractive index of the gas, ω is the laser frequency, and $\upsilon(\omega)$ is the ratio between the refractive indexes of the external (fused silica) and internal (gas) media.
- Thus, the incident radiation intensity profile as a function of the radial coordinate r is given by

$$I_0(r) = I_0\, J_0^2\,(2.405\ r/a\,) \qquad (8)$$

where I_0 is the peak intensity and Jo is the zero-order Bessel function (Marcatili et al., 1984).

Supercontinuum Generation in Hollow Fibers

- By applying the same mode on the complex propagation constant $\beta(\omega), \beta$ the real phase constant of Eq. (8) , and imaginary, $\alpha/2$ (field attenuation constant), parts of the propagation constant are given by:

$$\beta = \frac{2\pi}{\lambda}\left[1 - \frac{1}{2}\left(\frac{2.405\,\lambda}{2\pi\,a}\right)^2\right] \qquad (9a)$$

$$\frac{\alpha}{2} = \left(\frac{2.405}{2\pi}\right)^2 \frac{\lambda^2}{2\,a^3}\,\frac{\upsilon^2+1}{\sqrt{\upsilon^2-1}} \qquad (9b)$$

- where λ is the laser wavelength in the gas medium. Assuming Gaussian pulse profile and neglecting dispersion and self-focusing, the maximum broadening spectrum after propagating a length of l can be written as

$$\delta\omega_{max} = 0.86 \int_0^l \gamma\,(z) P_0\,\xi\, e^{-\alpha z} dz/T_0 \qquad (10)$$

- where z is propagating distance, α is given by Eq. (9b), P_0 is the peak power; T_0 is the half-width (at the 1/e intensity point) of the pulse; γ is the nonlinear coefficient and is given by $\gamma = n_2\; p(z)\,\omega_0/c\,A_{eff}$ [n_2 is given by Eq. (2)], where n_2 is the nonlinear index coefficient, ω_0 is the laser central frequency; c is the light speed in vacuum, ξ is the coupling efficiency, A_{eff} is the effective mode area (Nisoli et al., 1996). In the statically gas-filled case, the pressure is constant along the fiber. While in the differentially pumped case, the pressure is chosen to be a minimum (0 bar) at the entrance and gradually increases along the fiber. This leads to the pressure distribution

Supercontinuum Generation in Hollow Fibers

- $p(z) = \left[p_0^2 + \left(\frac{z}{L}\right)(p_L^2 - p_0^2)\right]^{1/2}$ (11)

- where p_0 and p_L are the pressure at the entrance and the exit, respectively. Then the bandwidth broadened in both the cases can be expressed as

- $\Delta\omega_{SF} = 0.86\omega_0 n_2\, p_L\, P_0\, \xi\, (1 - e^{-\alpha z})\, /\alpha\, c\, T_0\, A_{eff}\,,$ (12)

- $\Delta\omega_{DP} = 0.86\, \omega_0 n_2\, p_L \xi\, P_0 \int_0^l \frac{\sqrt{z}\, e^{-\alpha z}}{\sqrt{L}\, c\, T_0\, A_{eff}} dz.$ (13)

- So that the bandwidth broadening depends on both the input laser characteristics and the gas used in the hollow fiber. The nonlinear coefficient γ takes the values 7.4×10^{-25} m^2/W bar, 9.8×10^{-24} m^2/W bar, and 2.78×10^{-23} m^2/W bar for neon, argon, and krypton respectively (Nisoli et al., 1997, Robinson et al., 2006).

Our Modified Setup : *Ti:sapphire laser oscillator*

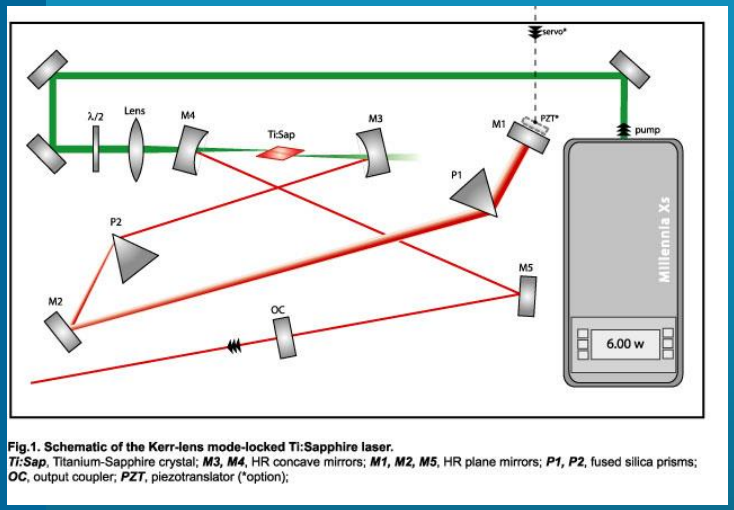
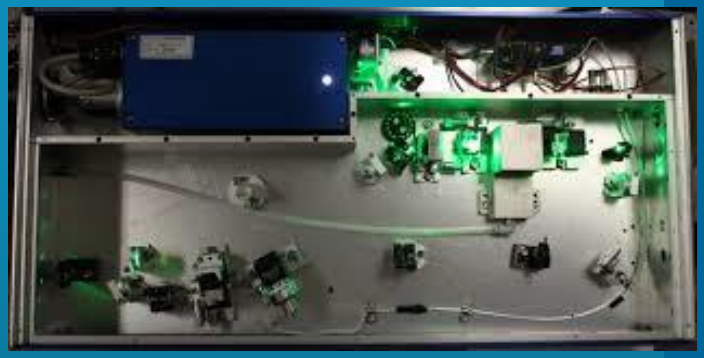
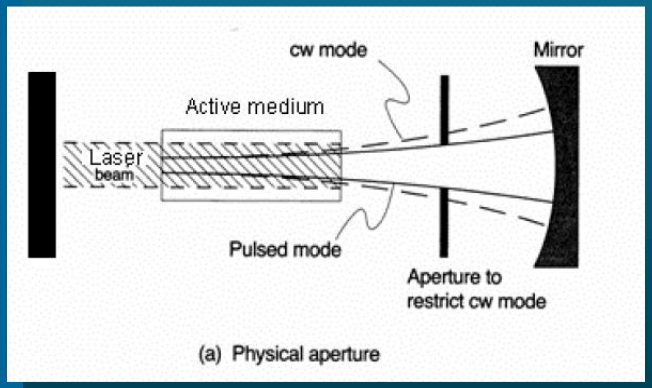
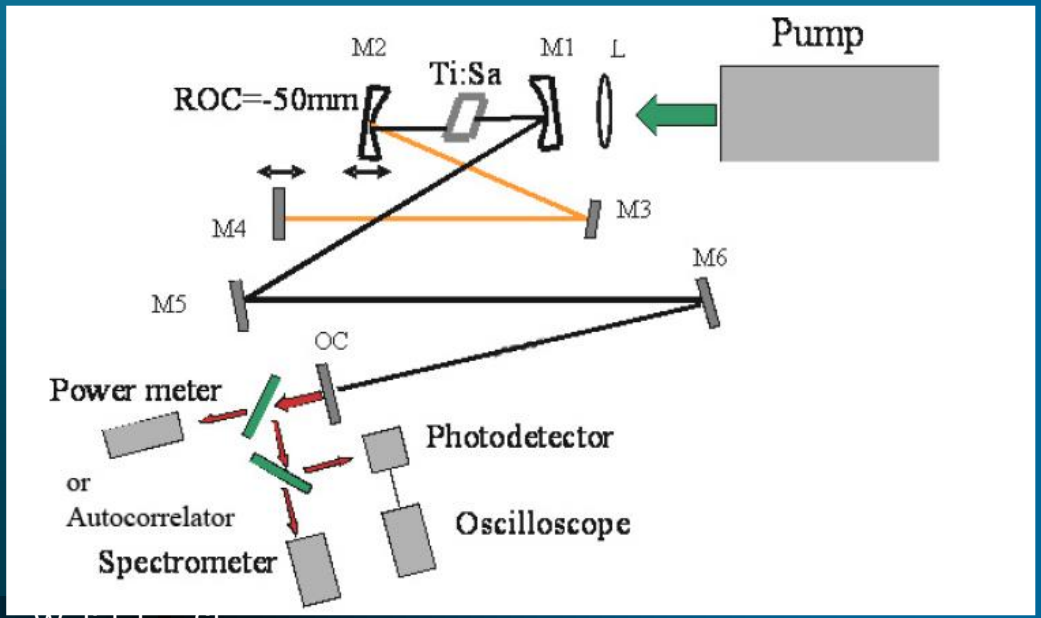


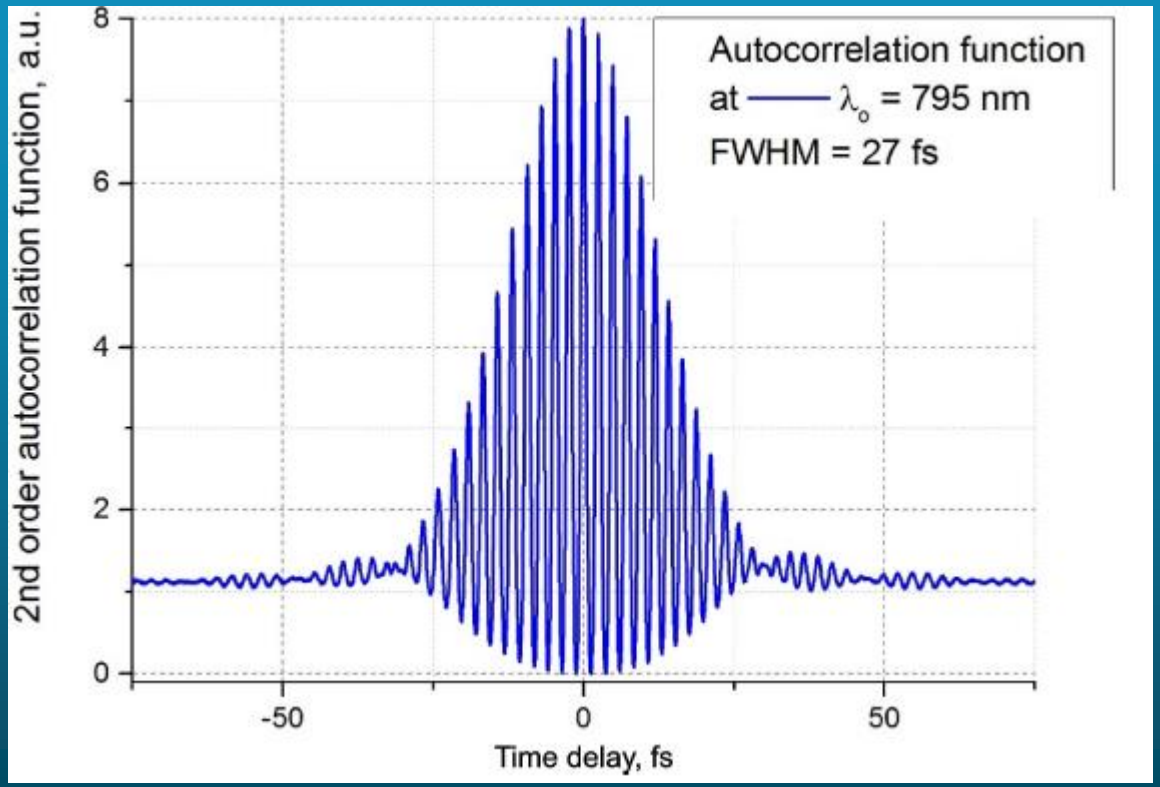
Fig.1. Schematic of the Kerr-lens mode-locked Ti:Sapphire laser.
Ti:Sap, Titanium-Sapphire crystal; ***M3, M4***, HR concave mirrors; ***M1, M2, M5***, HR plane mirrors; ***P1, P2***, fused silica prisms; ***OC***, output coupler; ***PZT***, piezotranslator (*option);





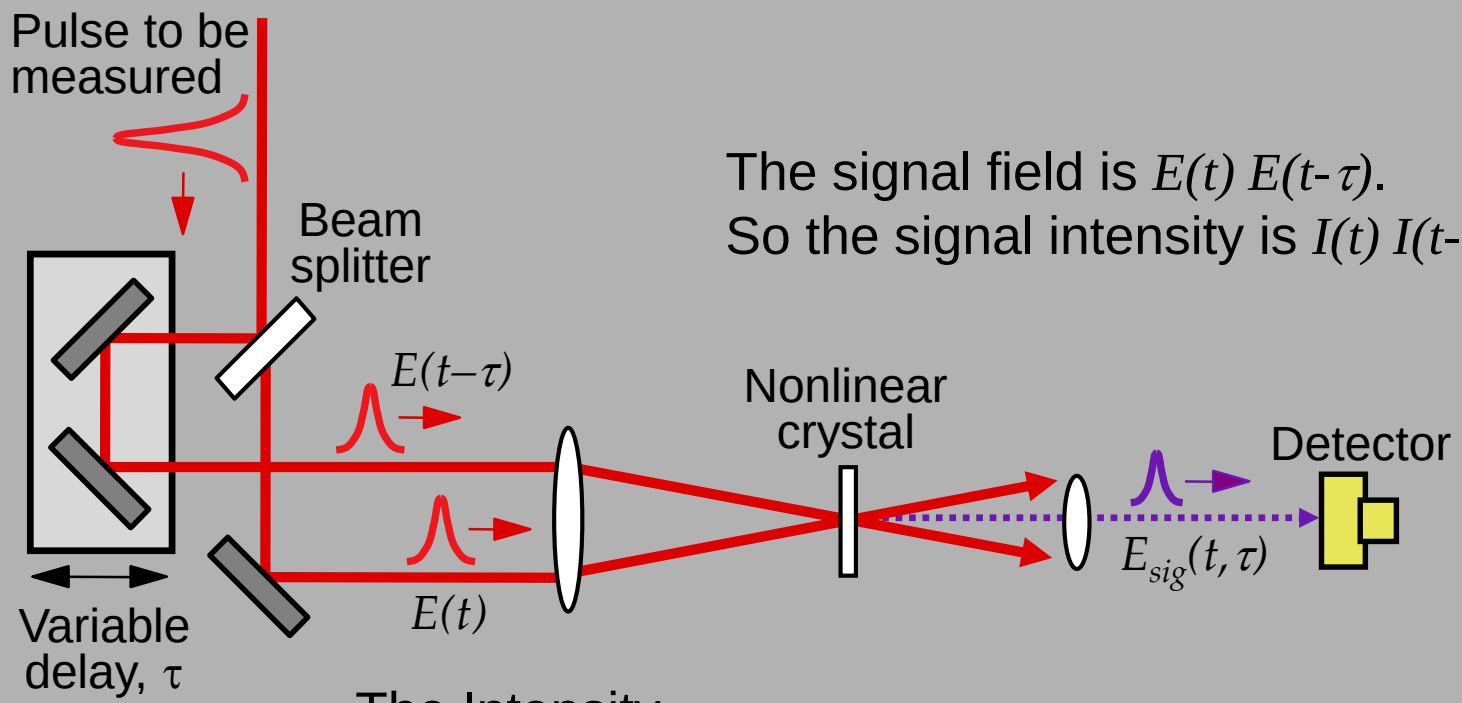


The 52 nm bandwidth of the Ti:sapphire oscillator at 795 nm m and 27 fs FWHM. **Gaussian Pulse**



Using the pulse to measure itself: The Intensity Autocorrelator

Crossing beams in a nonlinear-optical crystal, varying the delay between them, and measuring the signal pulse energy vs. delay, yields the **Intensity Autocorrelation, $A^{(2)}(\tau)$**.

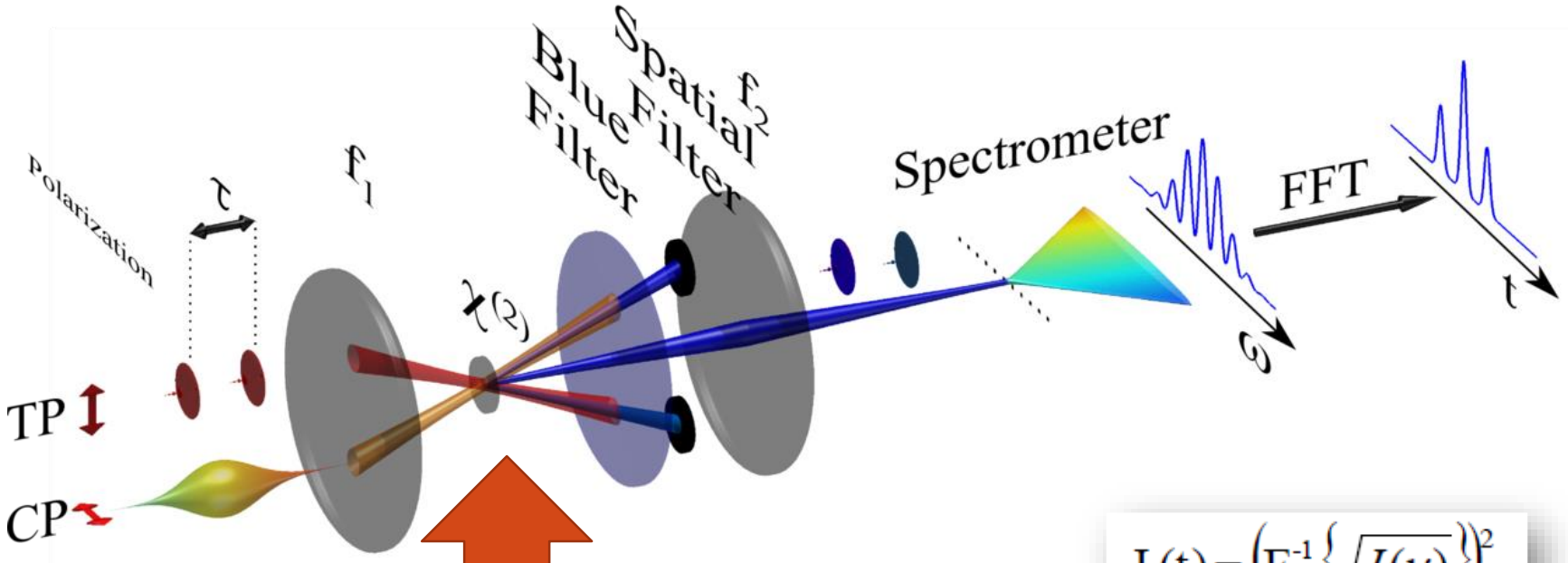


The signal field is $E(t)\,E(t-\tau)$.
So the signal intensity is $I(t)\,I(t-\tau)$

The Intensity Autocorrelation:

$$A^{(2)}(\tau) \equiv \int_{-\infty}^{\infty} I(t)I(t-\tau)dt$$

Concept of the spectral phase interferometry for direct electric-field reconstruction (SPIDER)

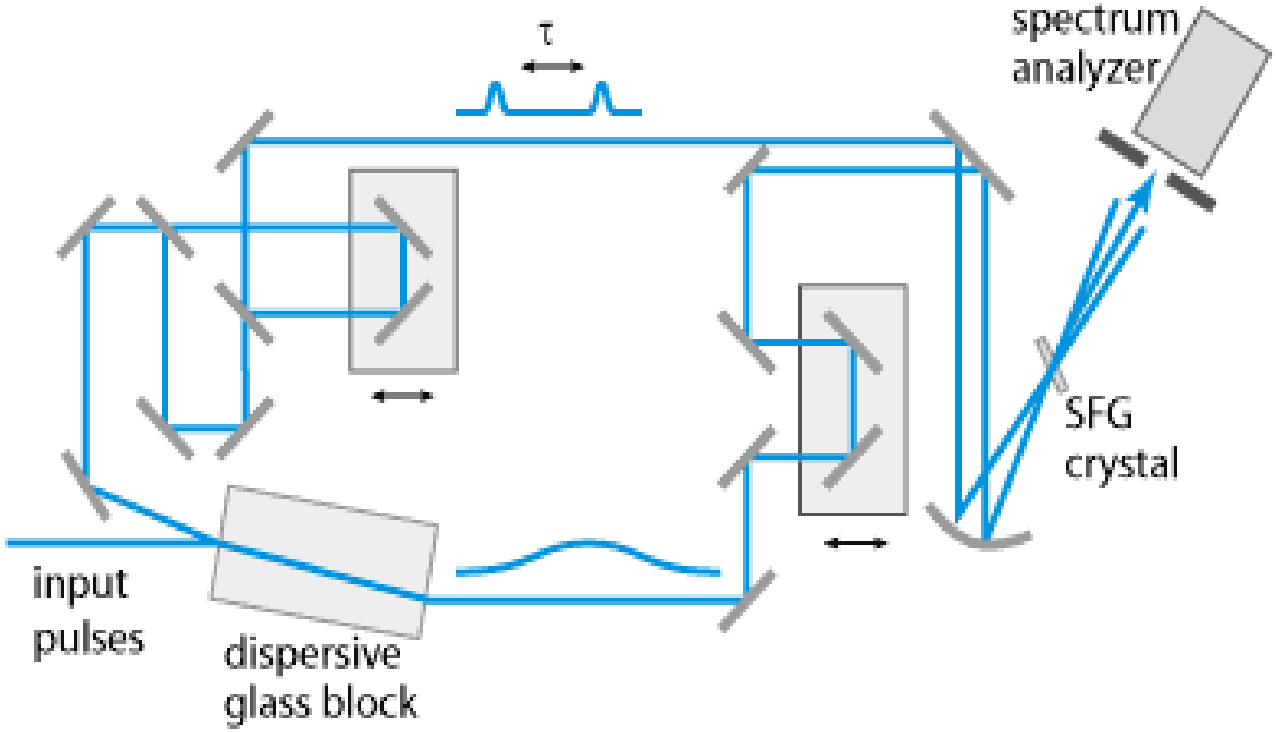


$$I(t) = \left(F^{-1}\left\{\sqrt{I(\nu)}\right\}\right)^2$$

Nonlinear frequency mixing

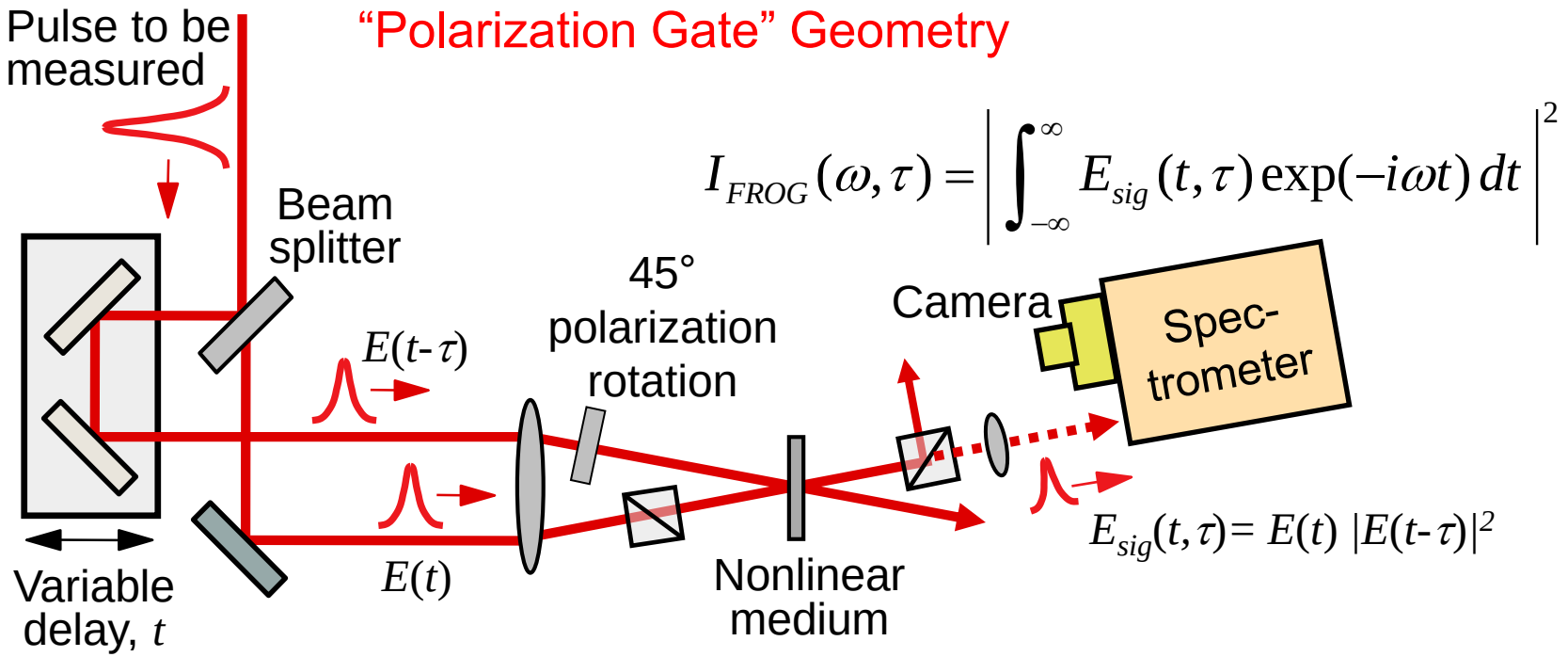
$$\varphi(\omega) = \omega\tau + \varphi_1(\omega) - \varphi_2(\omega)$$

Concept of the spectral phase interferometry for direct electric-field reconstruction (SPIDER)



Frequency-Resolved Optical Gating (FROG)

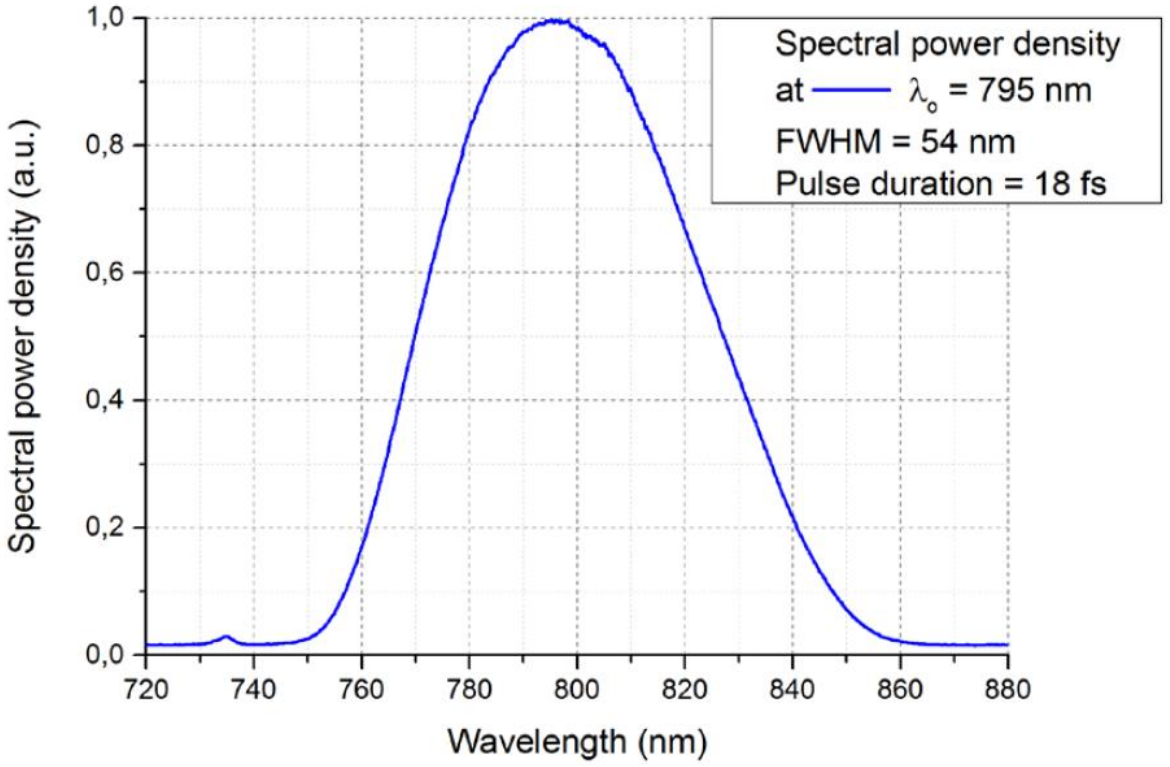
FROG involves gating the pulse with a variably delayed replica of itself in an instantaneous nonlinear-optical medium and then spectrally resolving the gated pulse vs. delay.



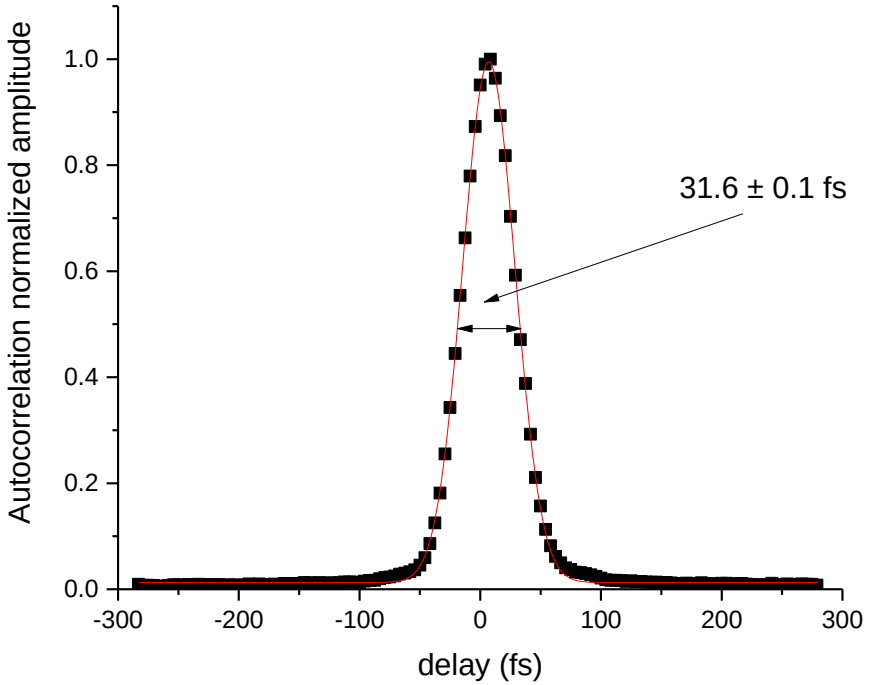
Use any ultrafast nonlinearity: Second-harmonic generation, etc.

R. Trebino, *Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses*, Kluwer

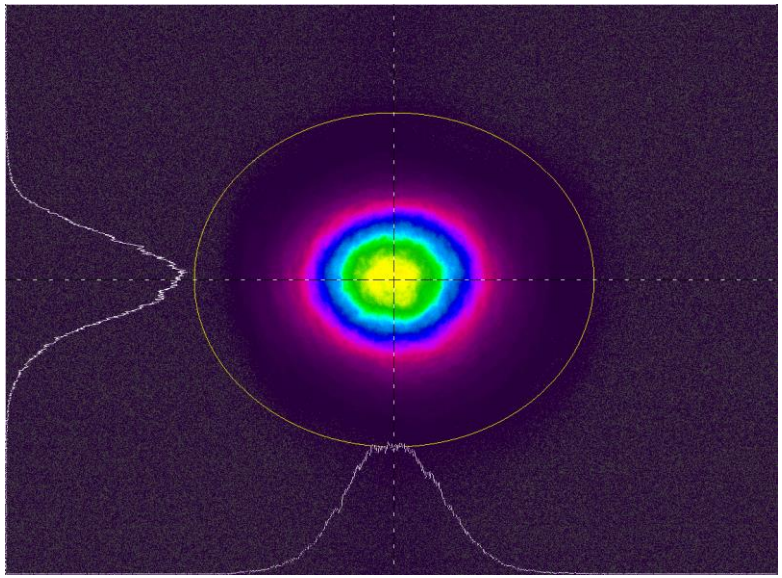
The 52 nm bandwidth of the Ti:sapphire oscillator at 795 nm at λ_0 = 795 nm and 18 fs pulse duration.



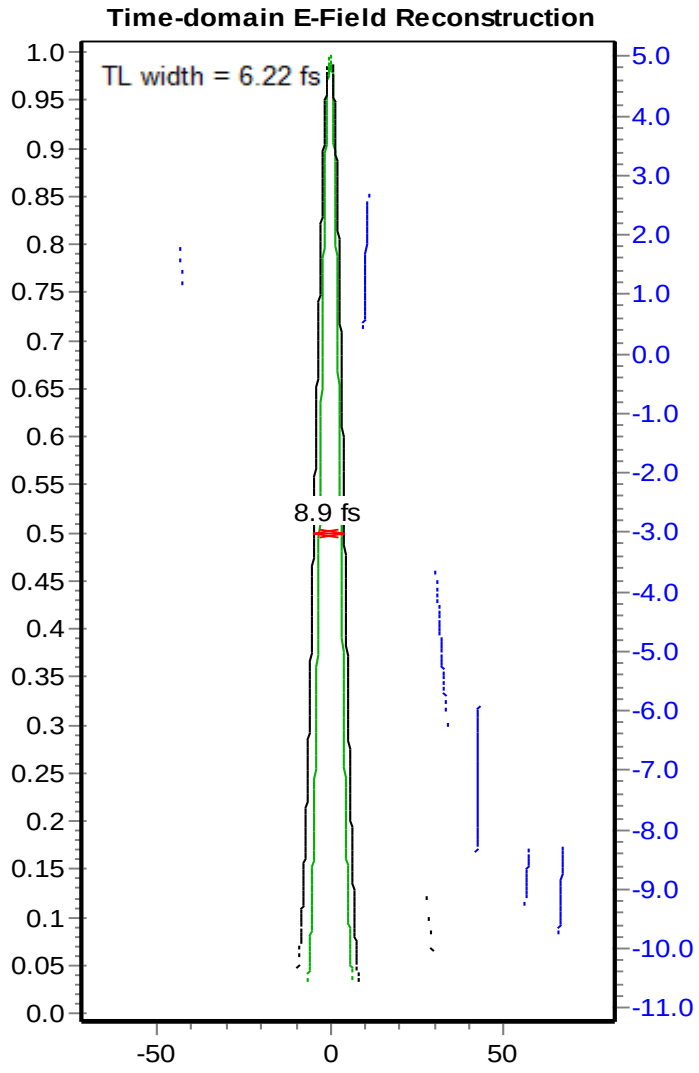
The autocorrelation measurement of the of input pulse duration of about 30 fs



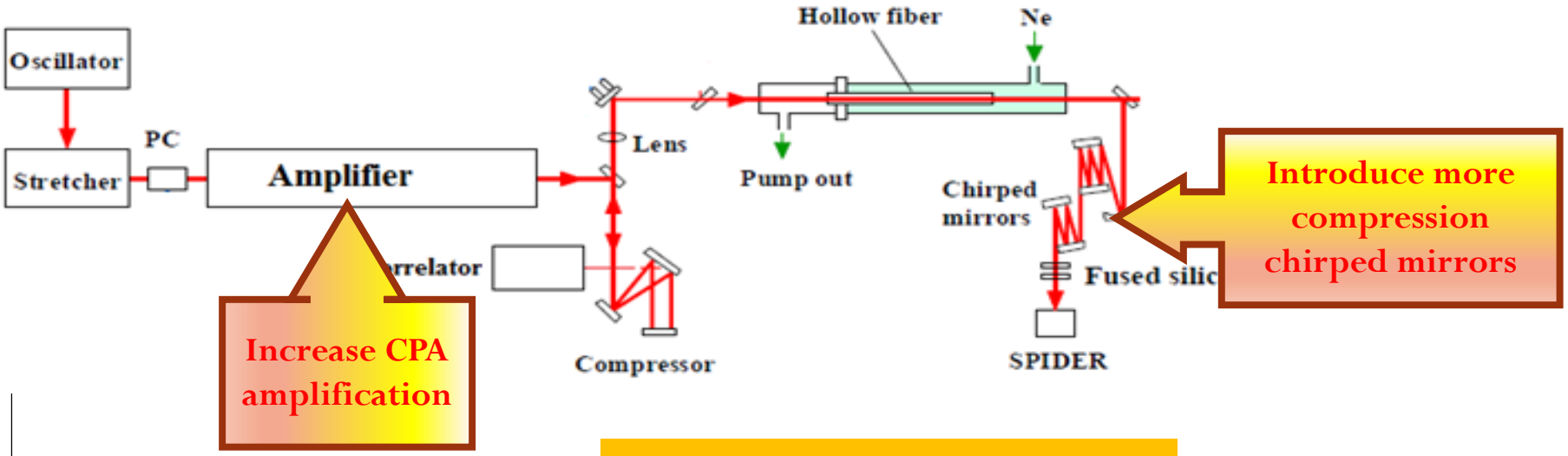
Beam profile images of 2.5 mJ output beam of CPA power amplifier, which shows TEM_{00} Gaussian transverse distribution.



The femtosecond output temporal profile after compression (green curve)



Our Modified experimental Setup

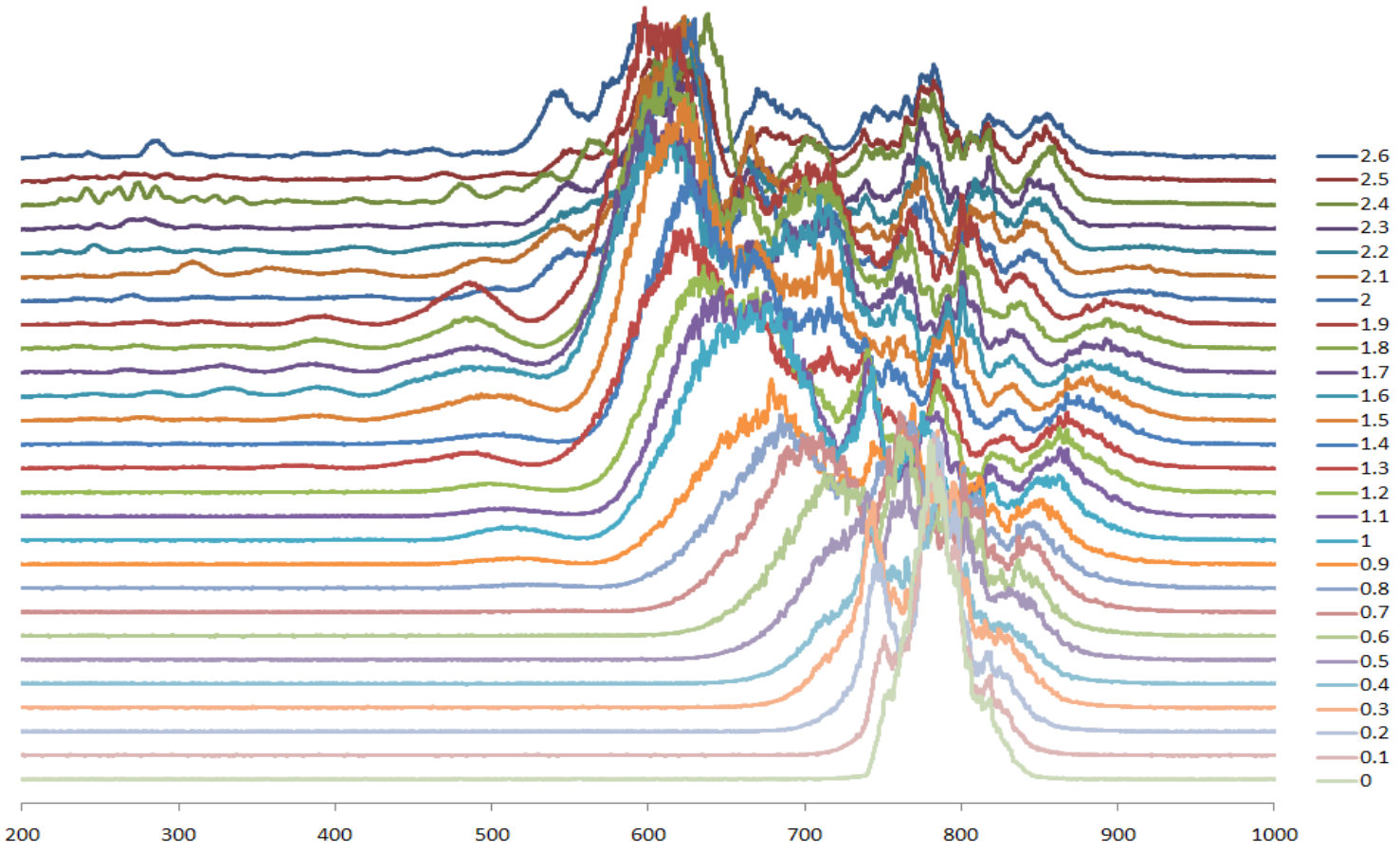


Increase number of chirped mirrors

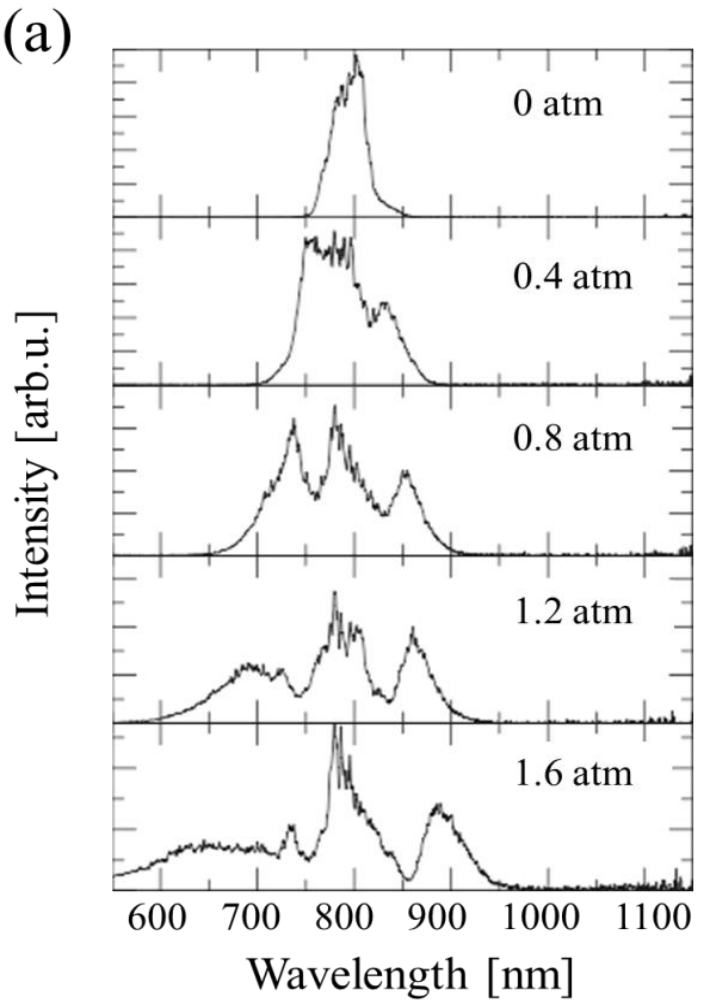
Increase pulse compression + Allow increasing CPA amplification

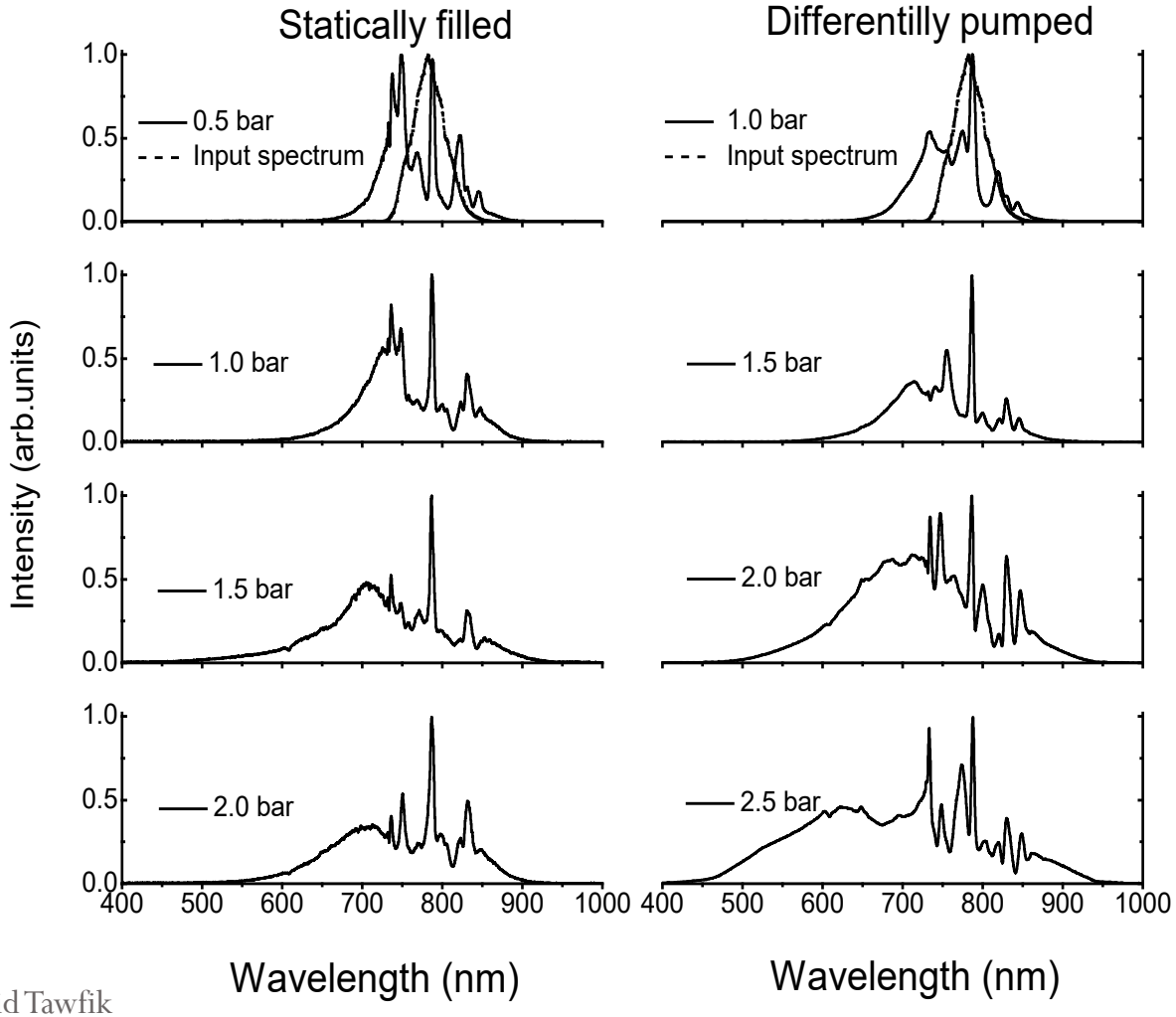
The pulse duration reaches **6.01 fs** and energy reaches **720 µJ**

Supercontinuum spectra vs Ne pressure (0 bar ~ 2.6 bar)

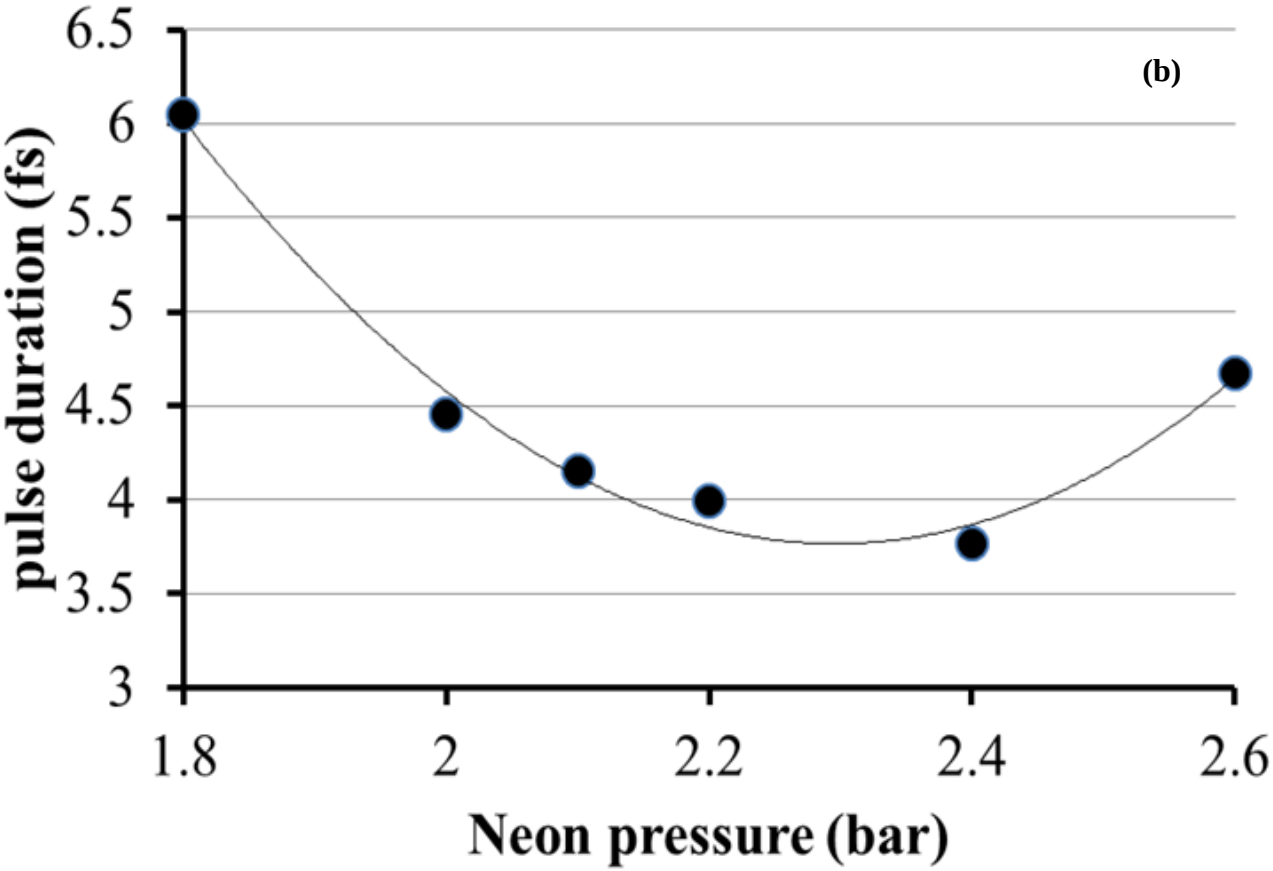


Supercontinuum spectra vs Ne pressure (0 bar ~ 2.6 bar)

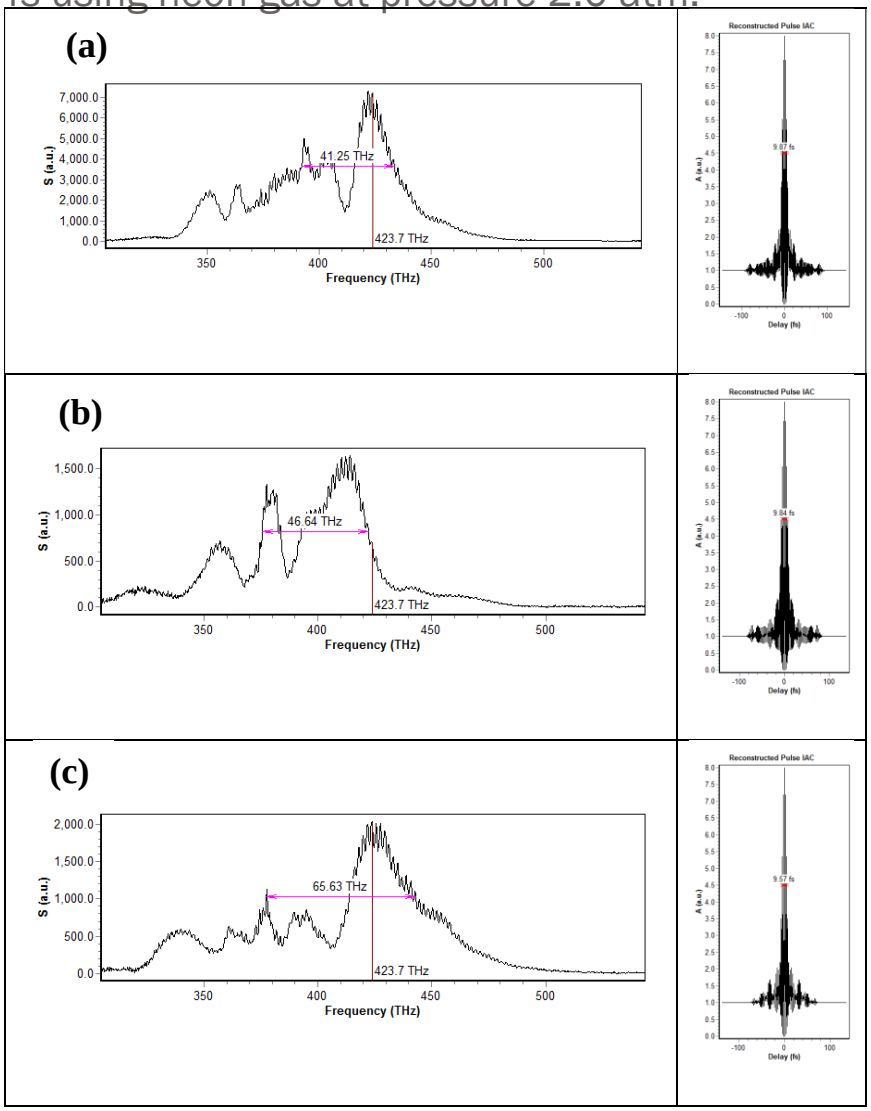



Statically filled
Differentilly pumped
Intensity (arb.units)
1.0
0.5
0.0
0.5 bar
Input spectrum
1.0 bar
Input spectrum
1.0 bar
1.5 bar
1.5 bar
2.0 bar
2.0 bar
2.5 bar
400
500
600
700
800
900
1000
Wavelength (nm)
Wavelength (nm)

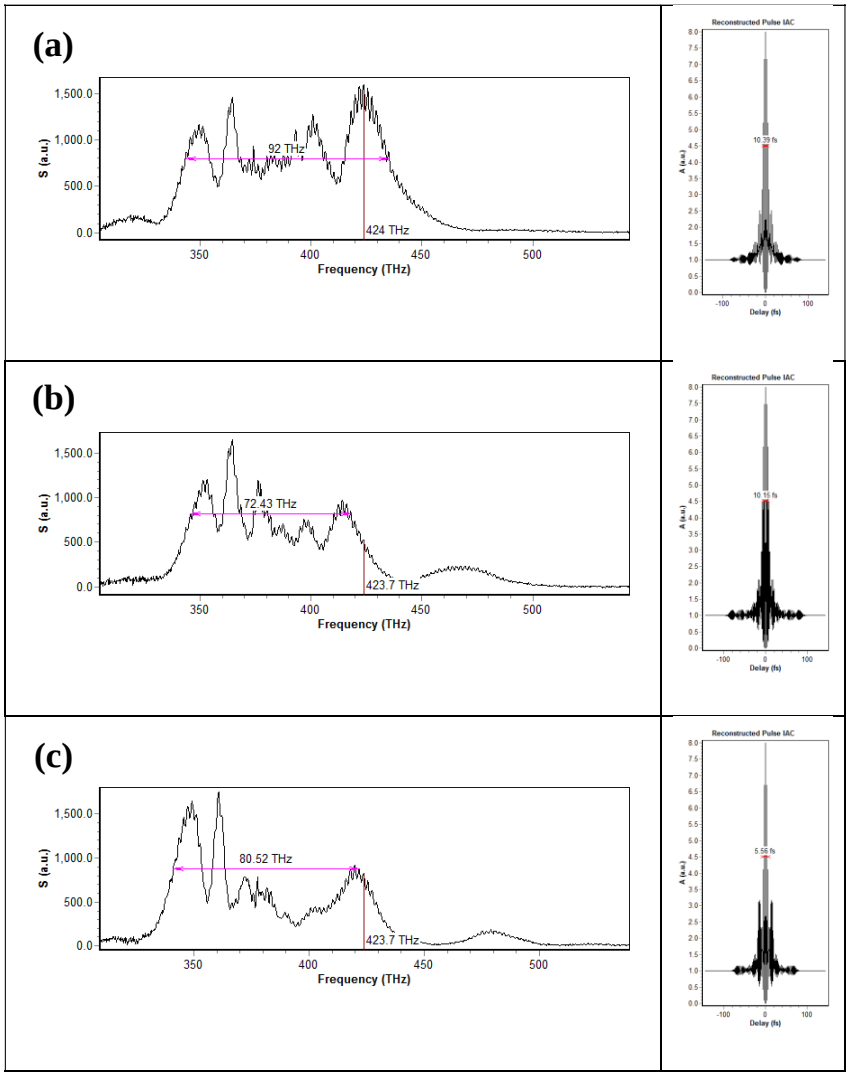
variations of the pulse duration with the neon gas pressure



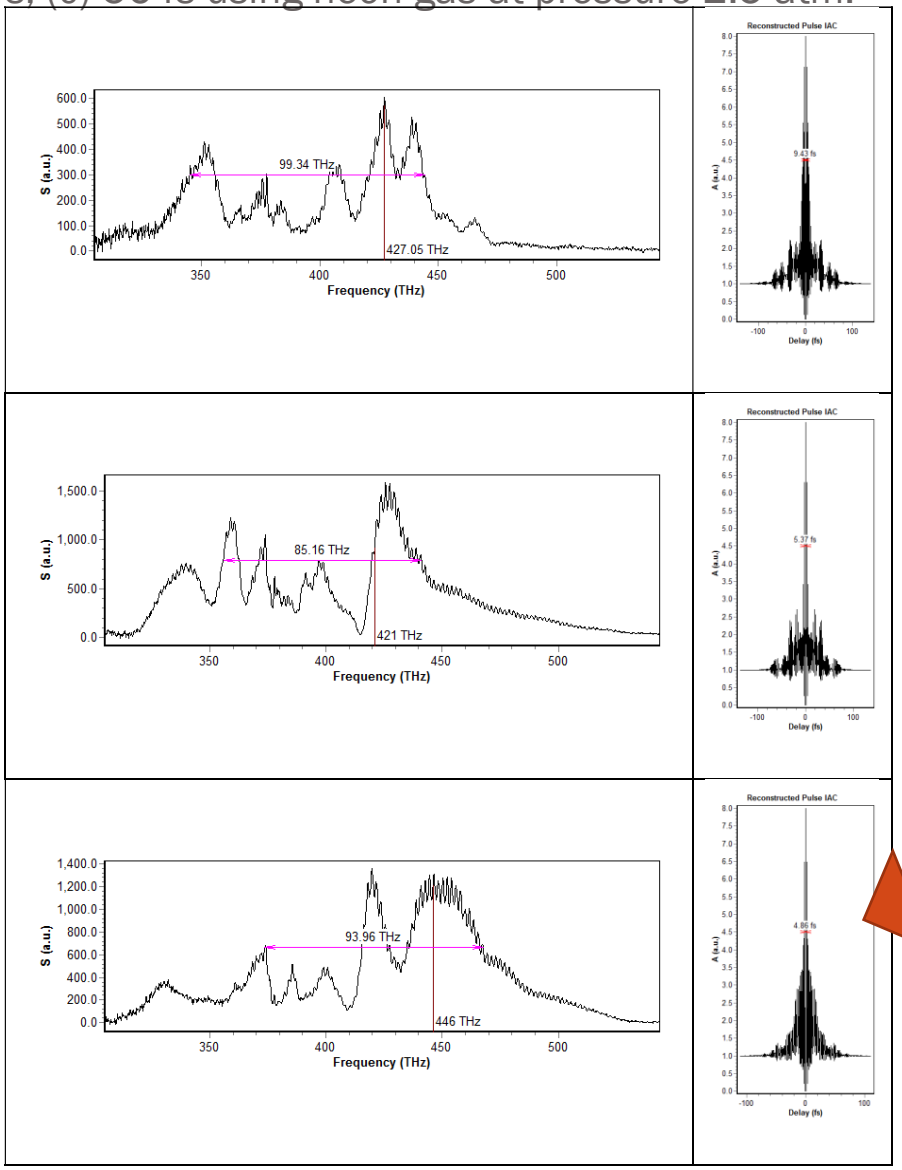
the output pulse spectral bandwidth broadening at the full-width-half-maximum (FWHM) (left) with the corresponding pulse duration (right) for the input pulse durations of (a) 32 fs; (b) 40 fs; (c) 56 fs using neon gas at pressure 2.0 atm.



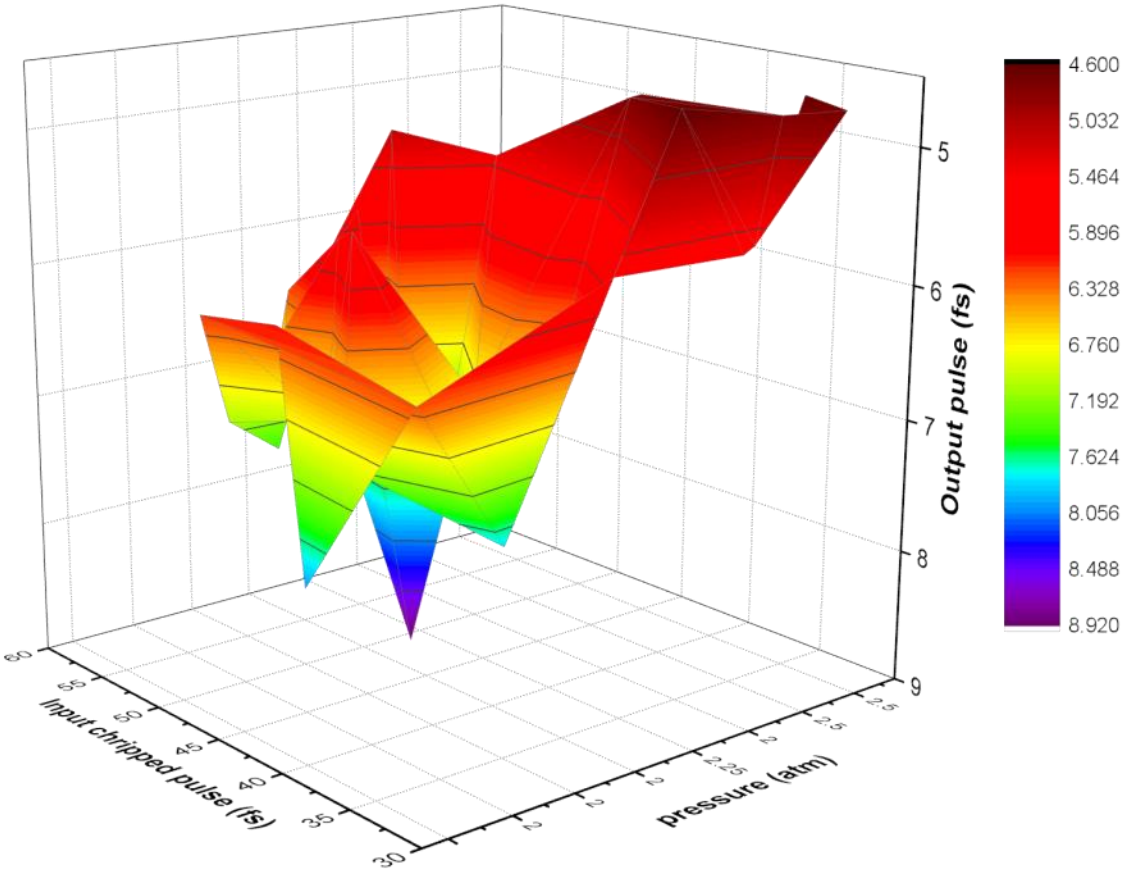
the output pulse spectral bandwidth broadening at the full-width-half-maximum (FWHM) (left) with the corresponding pulse duration (right) for the input pulse durations of (a) 32 fs; (b) 40 fs; (c) 56 fs using neon gas at pressure 2.25 atm.



the output pulse spectral bandwidth broadening at the full-width-half-maximum (FWHM) (left) with the corresponding pulse duration (right) for the input pulse durations of (a) 32 fs; (b) 40 fs; (c) 56 fs using neon gas at pressure 2.5 atm.



3D representation of the observed femtosecond temporal profile changes for different ICP values and neon gas pressures. The color pattern describes the output pulse from most compressed output pulse (red) to the low compressed pulse (violet) .



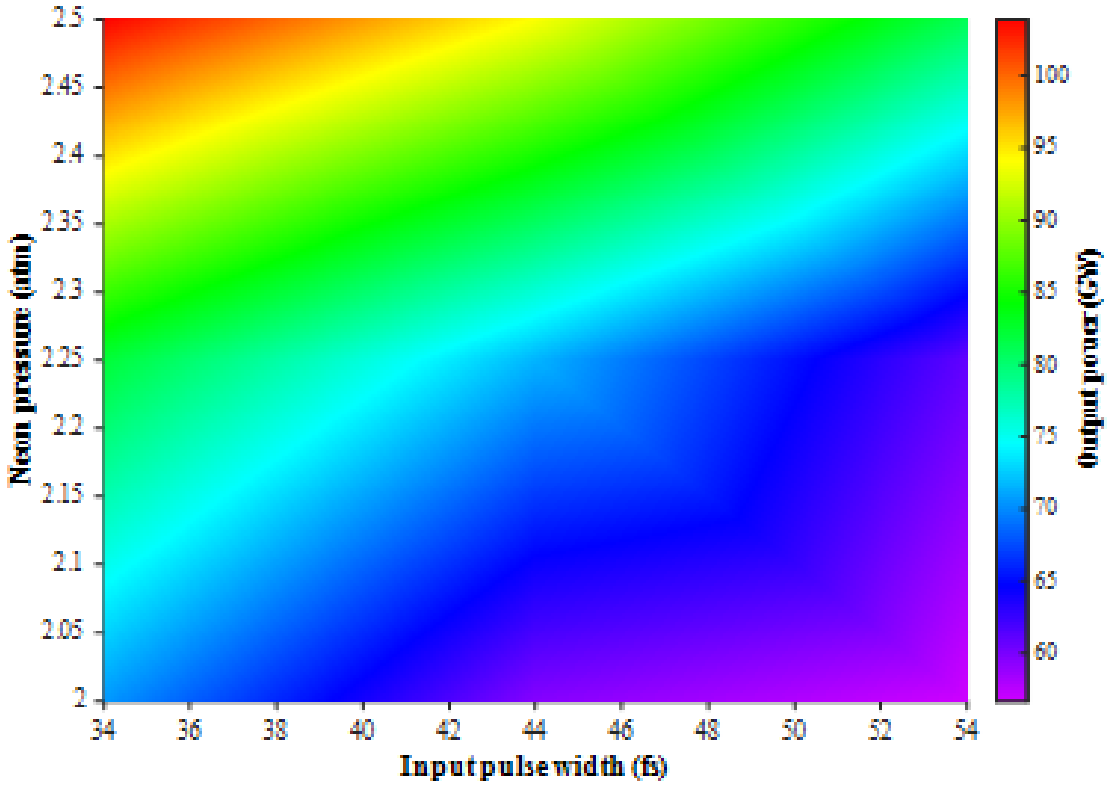
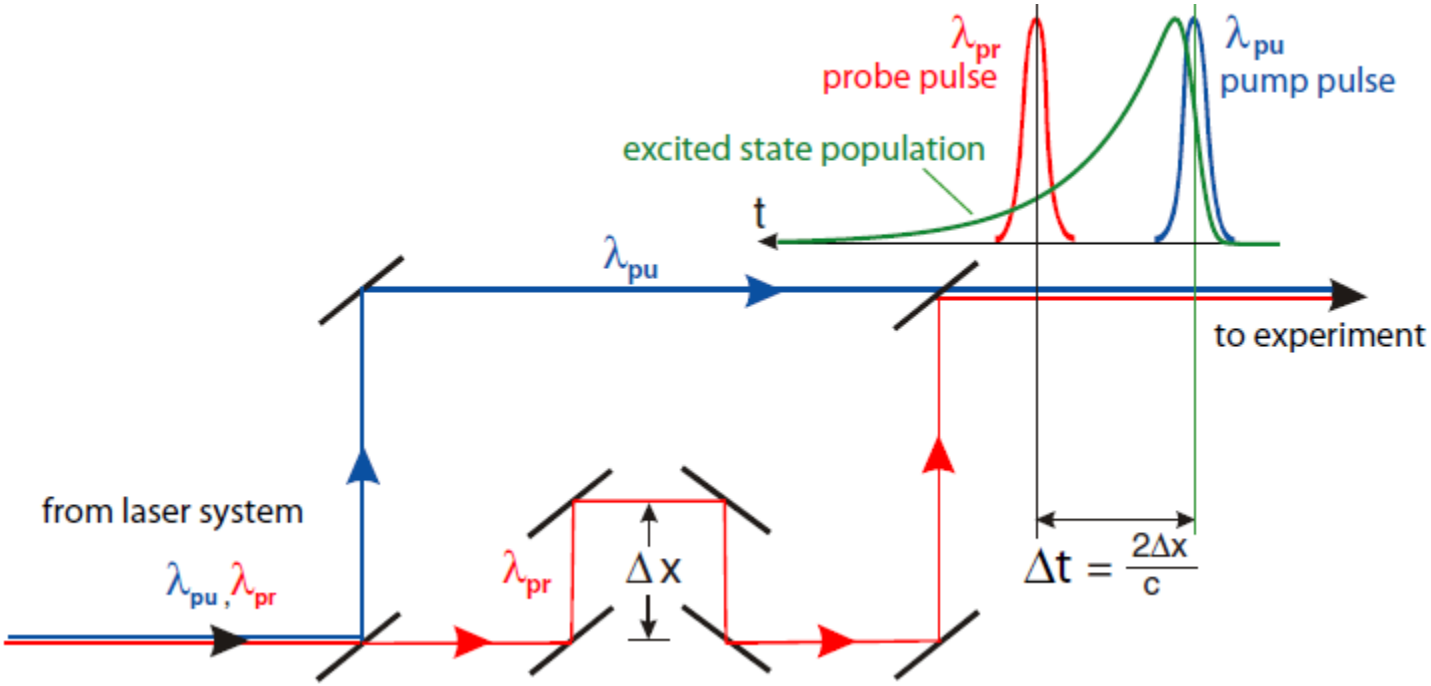


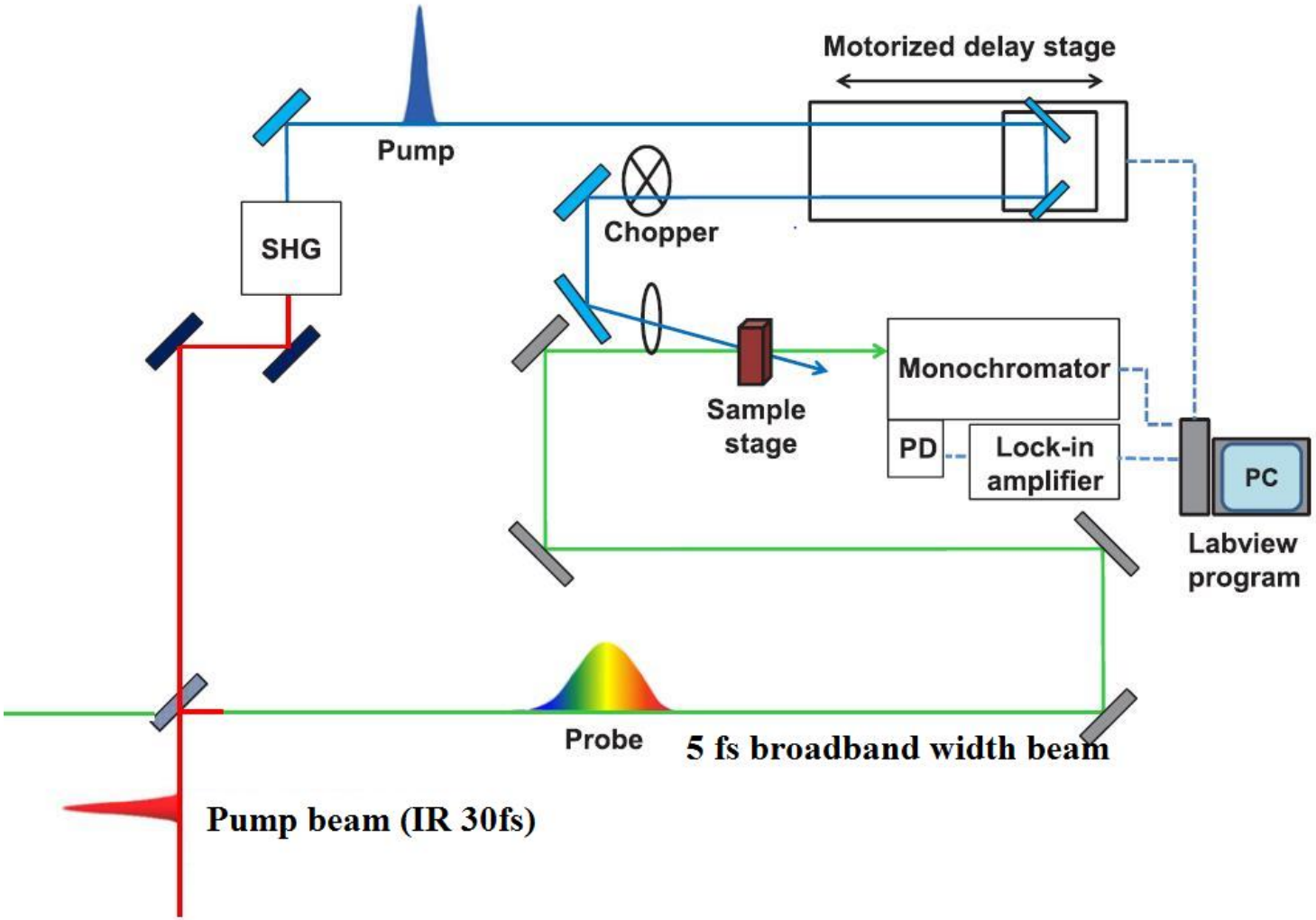
Fig.3 A summarized 2D contour representation of time-domain electric field reconstructed pulse profile of the observed TLPPP values from 56.7 to 103.9 GW for different τ_i values in the range from 32 to 54 fs, and neon gas pressures from 2-2.5 atm.

Future prospective pump-probe experiment for complex molecules



Schematic of the pump–probe concept for detection of ultrafast processes. The pump pulse (wavelength λ_{pu}) excites the molecular (atomic) system which then evolves with time *t* . Its population may, e.g. decay exponentially. The probe beam hits the target after a delay time *t* and transfers it into a state which can be directly monitored experimentally. The delay time *t* can be varied by an optical delay line **Rep. Prog. Phys. 69 (2006) 1897–2003 .**

Future prospective pump-probe experiment for complex molecules




Thanks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