Picosecond radiolysis and photolysis at Saclay

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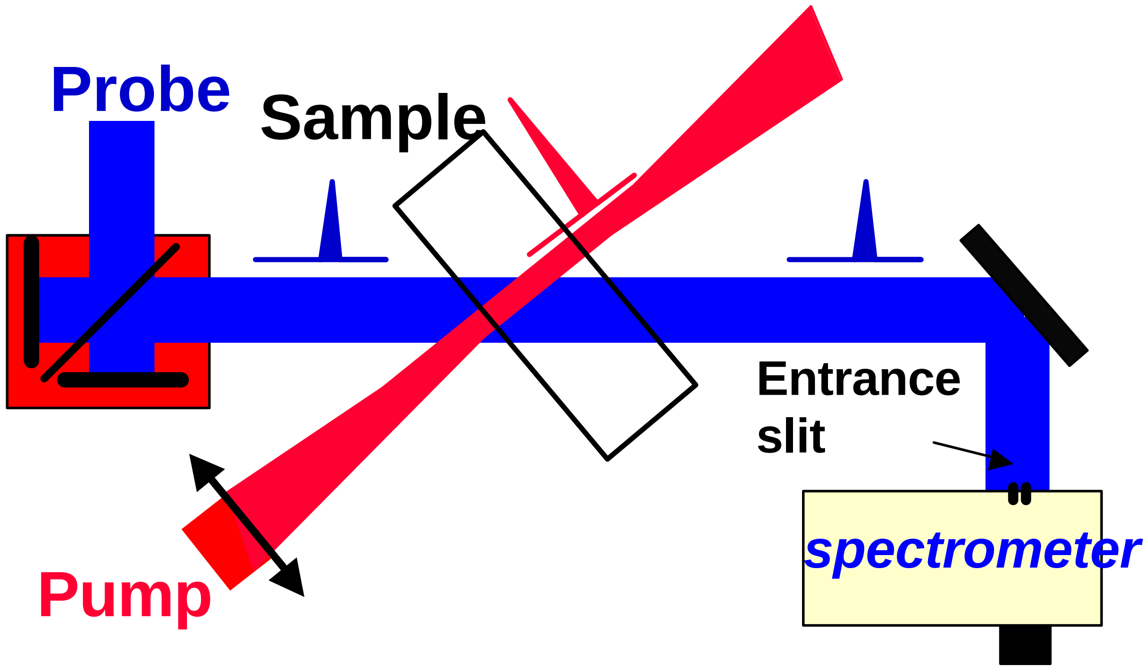
Why CEA should care about subpicosecond radiolysis?

- Highly concentrated media
  - Fuel recycling
- Nanostructured media
  - Waste management
- High Temperature High Pressure
  - Generation IV

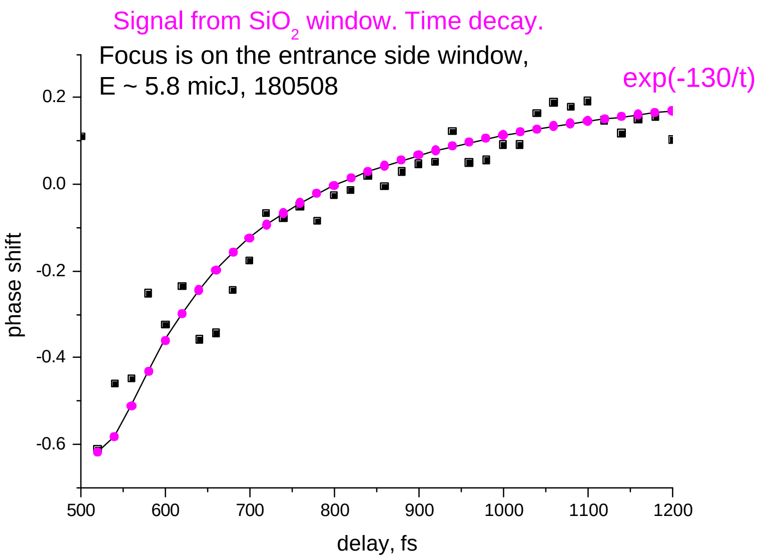
- Increasing the knowledge on track chemistry
  - Influence of the particle on the picosecond chemistry
  - Influence of the particle on the reactivity prior to solvation
  - (Easy) to implement highly non linear time resolved nonlinear optical spectroscopy (SFG, NOPA, NIR et FIR, THz, …)

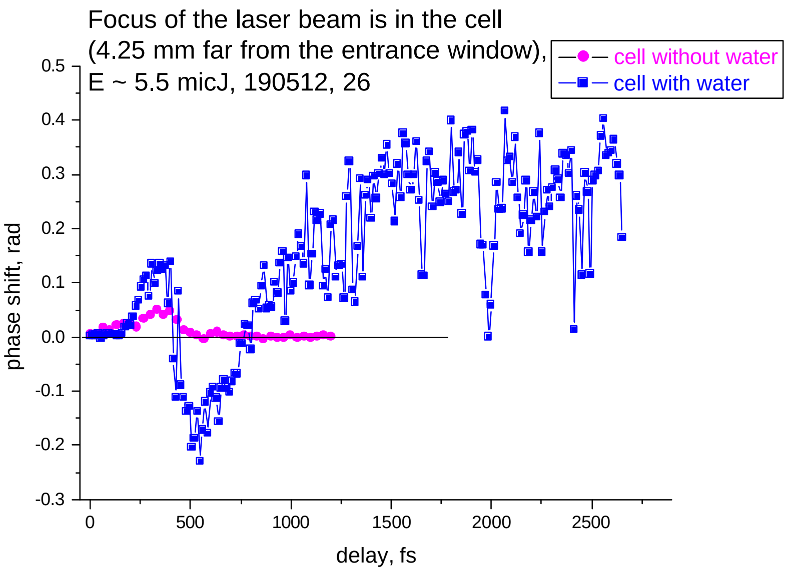
EXPERIMENTAL SET UP

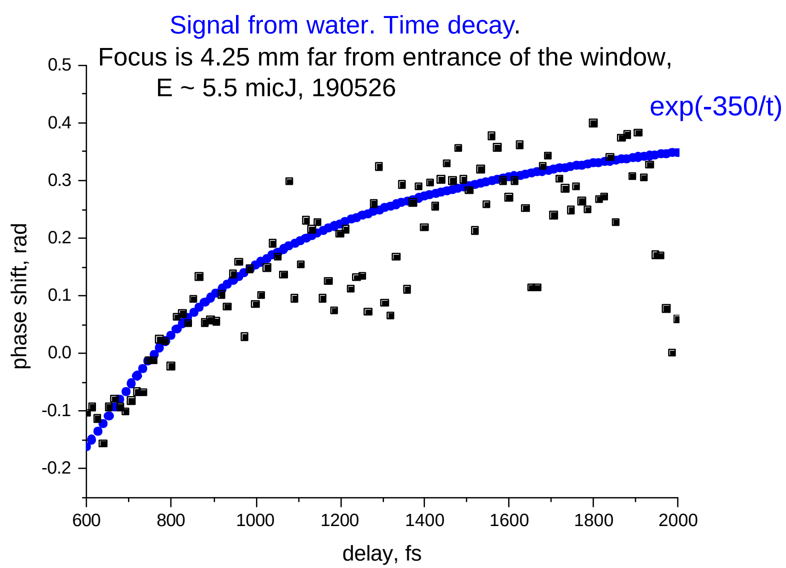


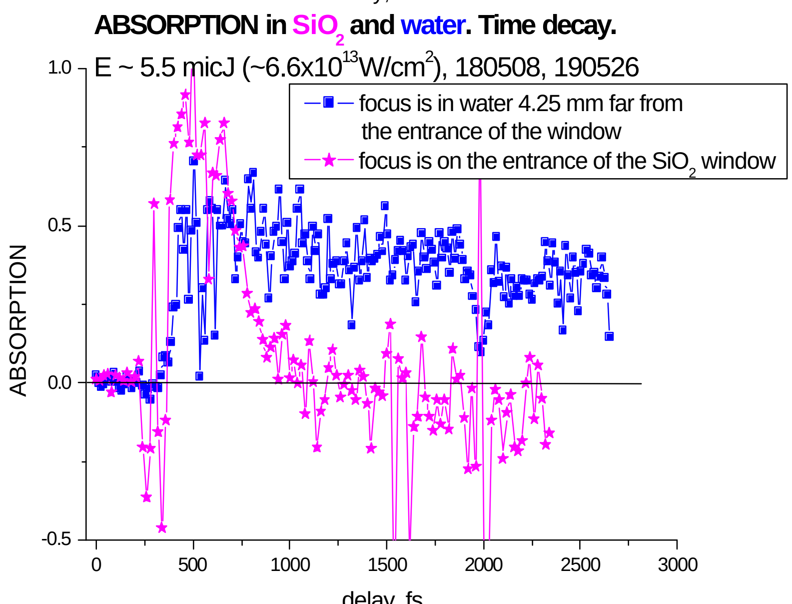
The sample is a cell 10 mm thickness with SiO_2 windows (with or without water).
Wavelength of the pump beam is 400nm.
Wavelength of the probe beam is 800nm
2 positions of the focus of the pump beam:
a) on the entrance side of the entrance window of the cell – to check a signal from SiO_2
b) 4.25 mm far (in a cell) – to check a signal from water

Comparison of signals with and without water







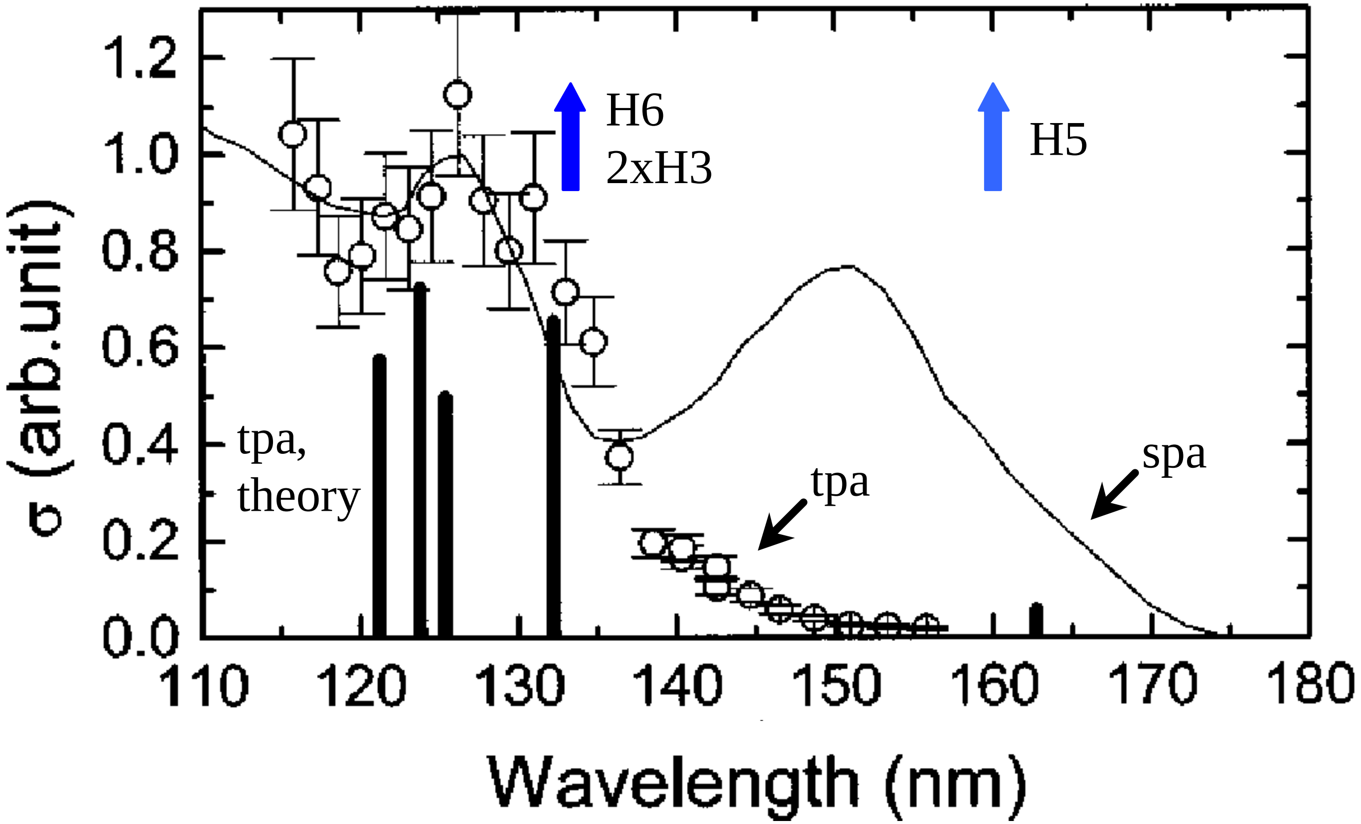


Femtosecond one-photon ionization of liquid water

- What is the quantum yield for the production of the hydrated electron ?
- What is the branching ratio between the ionization and the dissociation channels in the ´low´ energy ($E_{photon} < 12.6$ eV) photoexcitation of liquid water ?
- What is the thermalization distance of the ejected electron ?
- How do these quantities vary with excitation energy ?
- Is there any difference between single- and multi-photon excitation of identical or similar (total) energy (e.g. H6 vs. 2xH3) ?

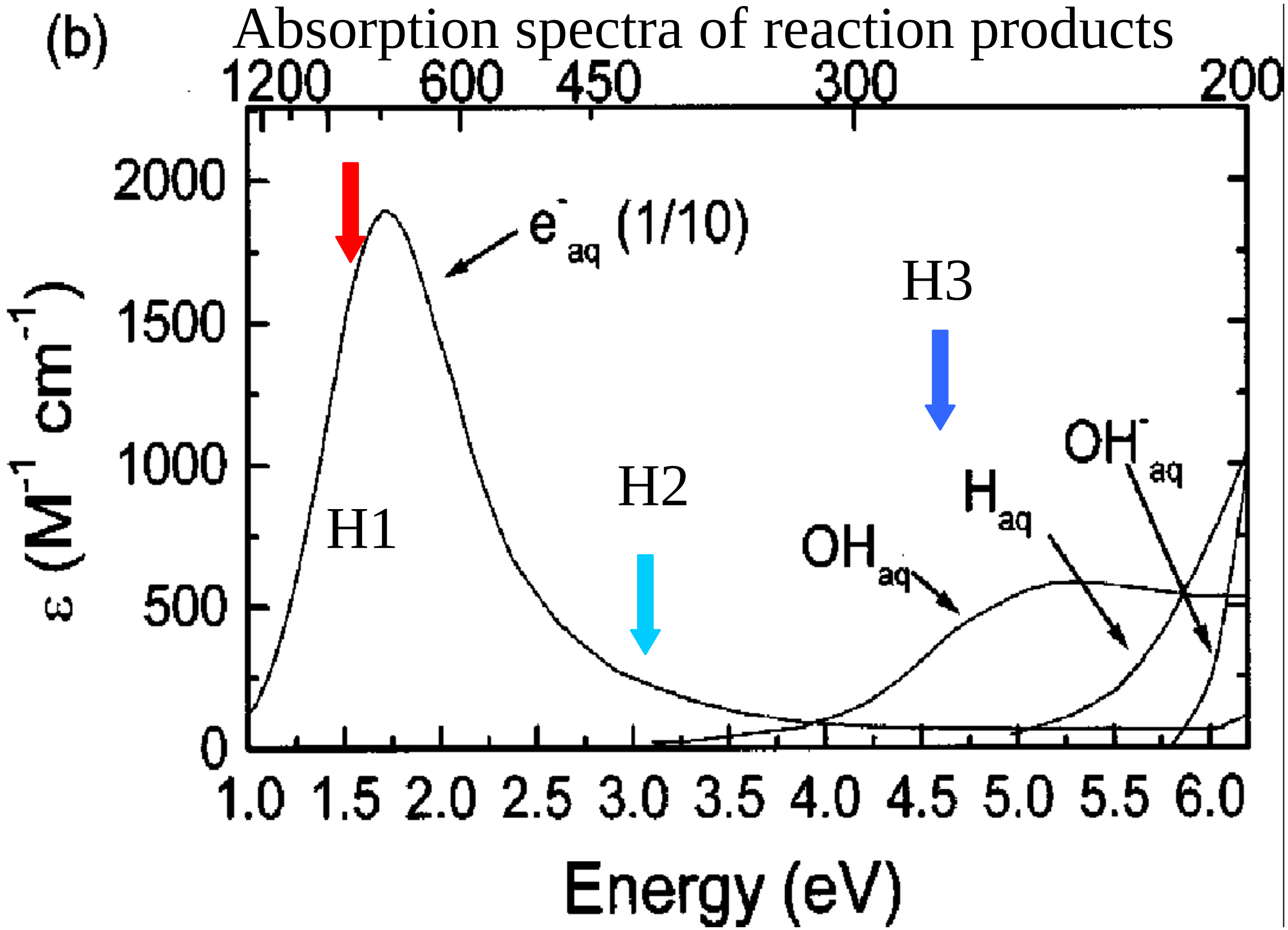
Femtosecond pump-probe transient absorption spectroscopy: the pump

A comparison between excitation with H5, H6 and 2xH3 is particularly interesting because of the erratic behavior of the one- and two-photon absorption cross sections of liquid water in this wavelength range:



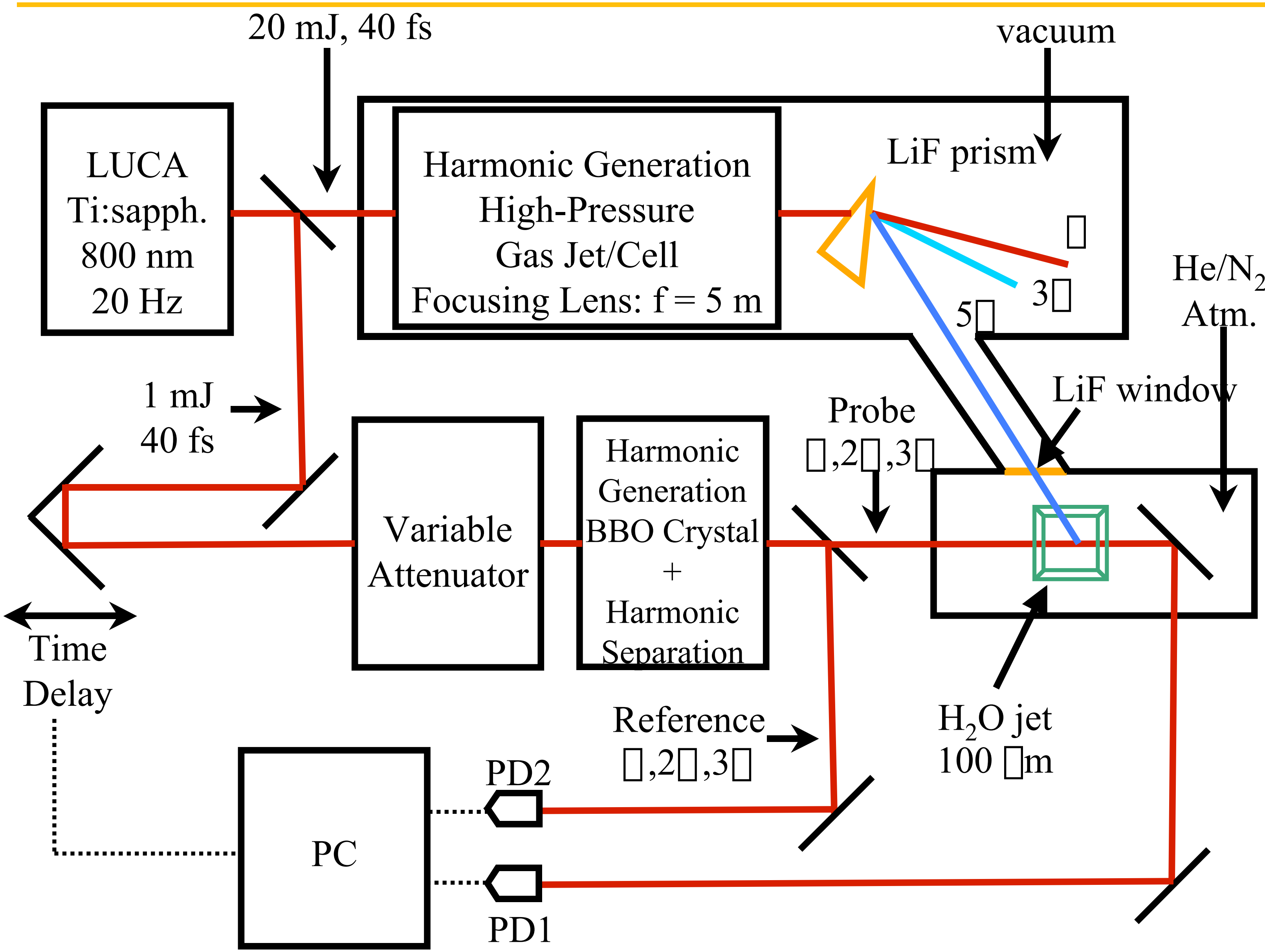
C.L. Thomsen *et al.* JCP **110** (7) 3453 (1999)

Femtosecond pump-probe transient absorption spectroscopy: the probe



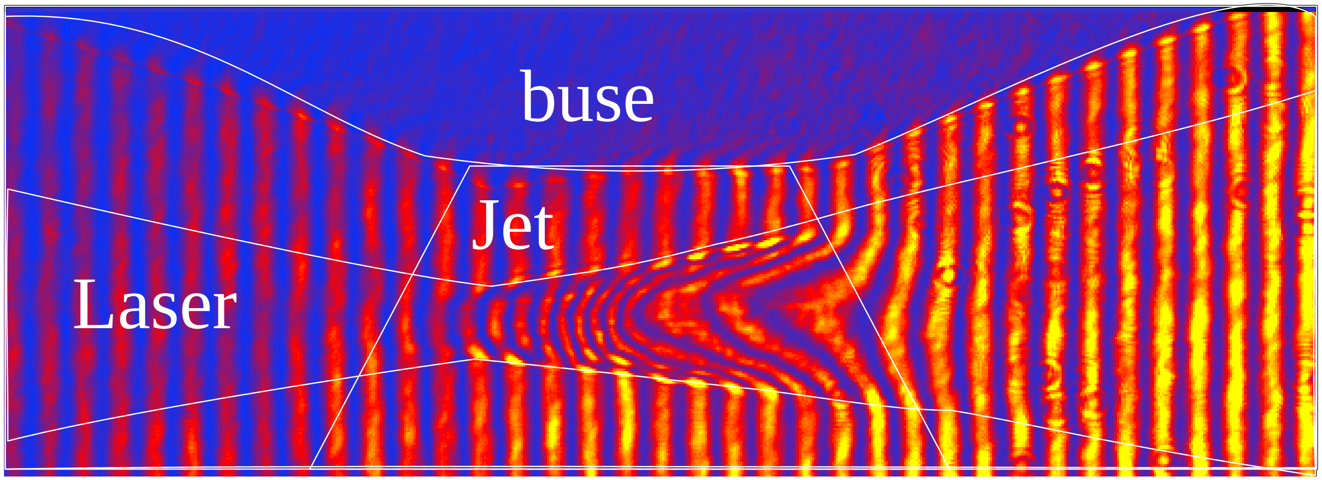
- Probe using the fundamental (e^-_{aq}), and the 2nd (e^-_{aq}) and 3rd (e^-_{aq} and OH_{aq}) harmonics of the Ti:sapphire laser

(b) C.L. Thomsen *et al*. JCP **110** (7) 3453 (1999)

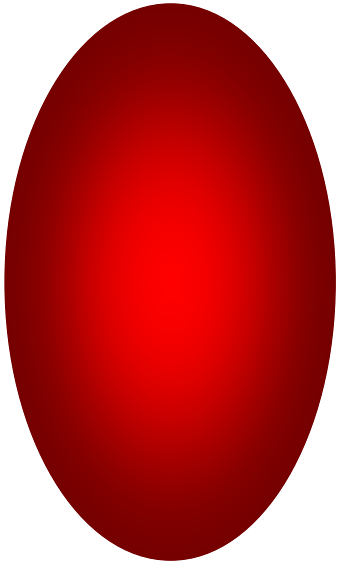

20 mJ, 40 fs
vacuum
LUCA
Ti:sapph.
800 nm
20 Hz
Harmonic Generation
High-Pressure
Gas Jet/Cell
Focusing Lens: f = 5 m
LiF prism
He/N_2
Atm.
LiF window
1 mJ
40 fs
Probe
Variable
Attenuator
Harmonic
Generation
BBO Crystal
+
Harmonic
Separation
Time
Delay
Reference
H_2O jet
PC
PD2
PD1

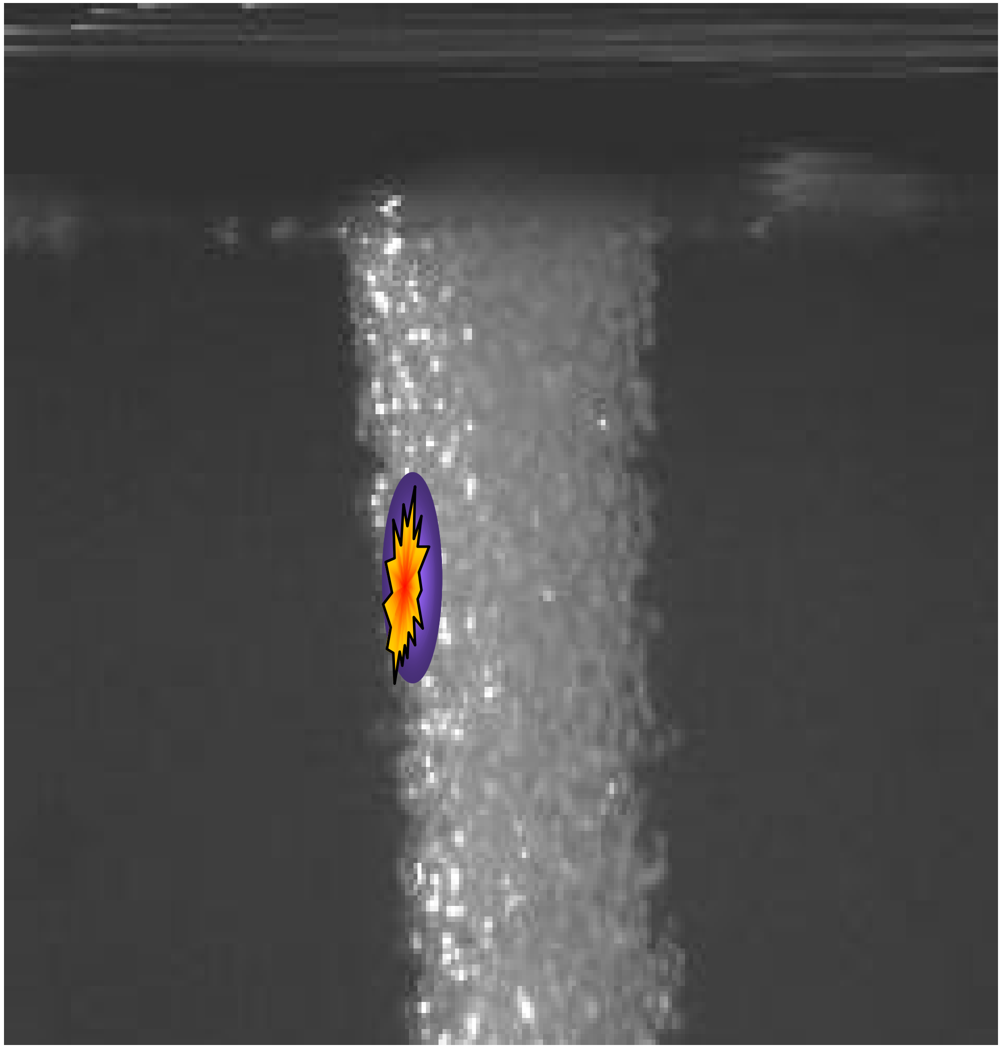
Laser-plasma interaction

Determination of the plasma density

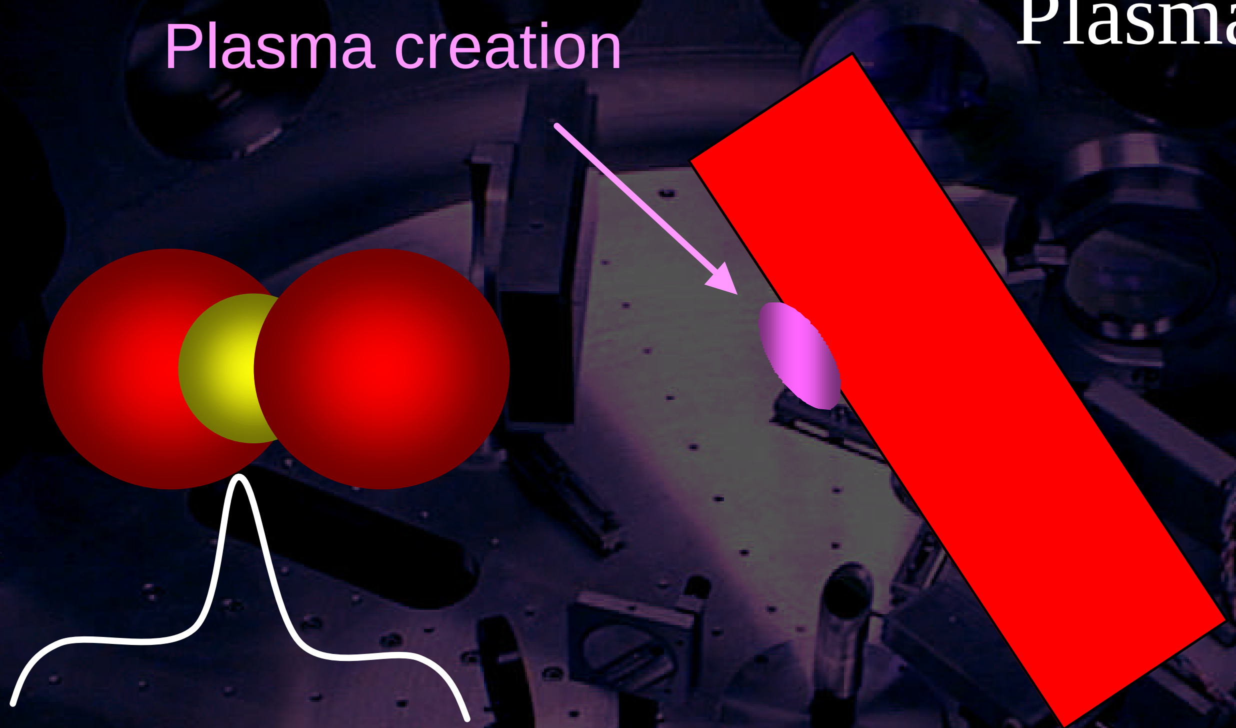


Génération de rayonnement X incohérent par focalisation sur des poudres



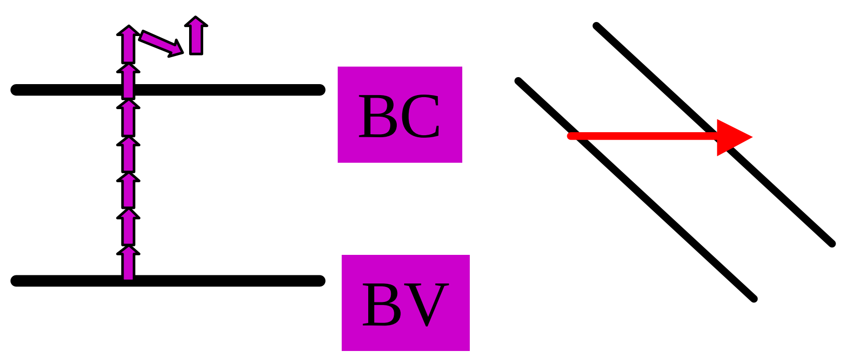


Plasma Miroir

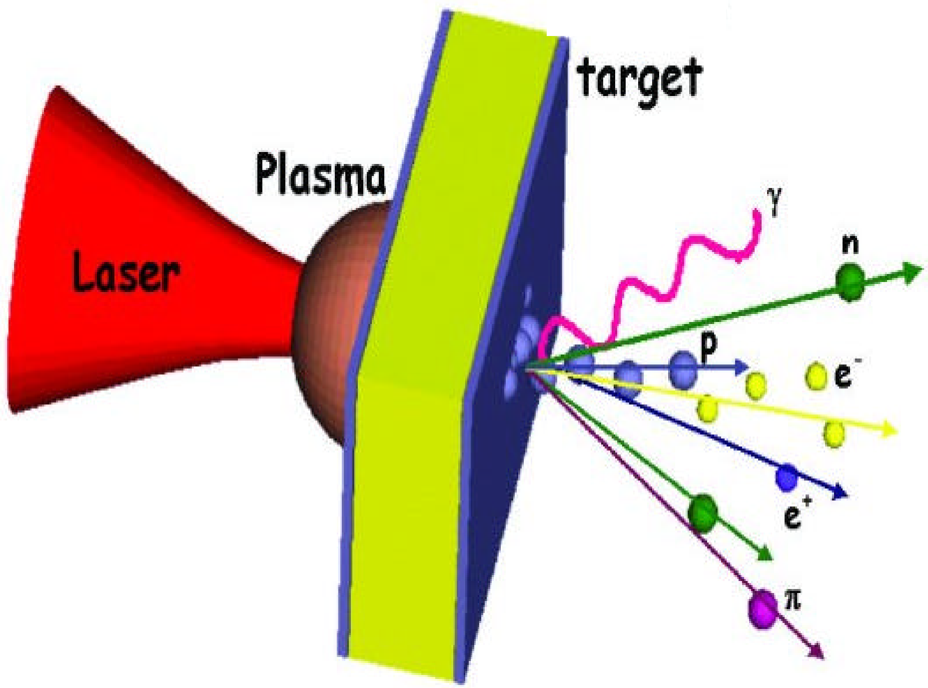


High electronic density in the conduction band:
dielectrique→metal
transparent→reflective

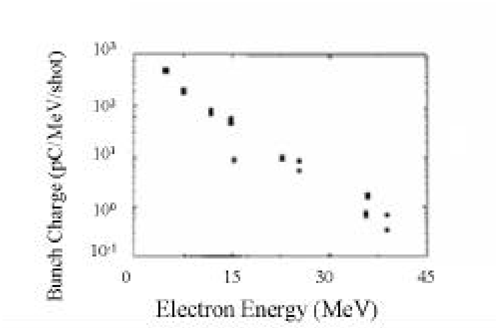
Principe physique
Ionisation Multiphotonique ou tunnel



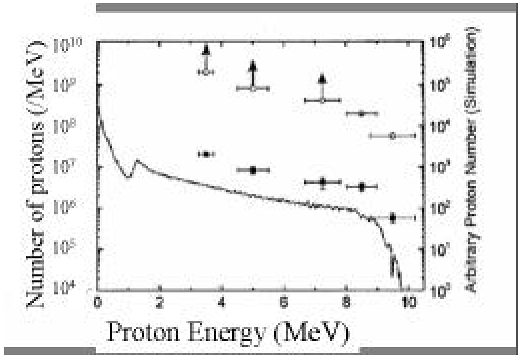
2005: Genaration of secondary particles in an controlled area.



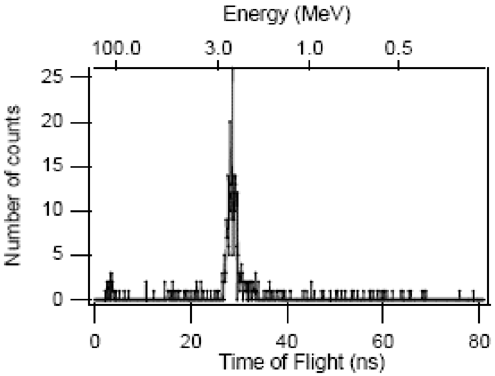
Electrons



Protons



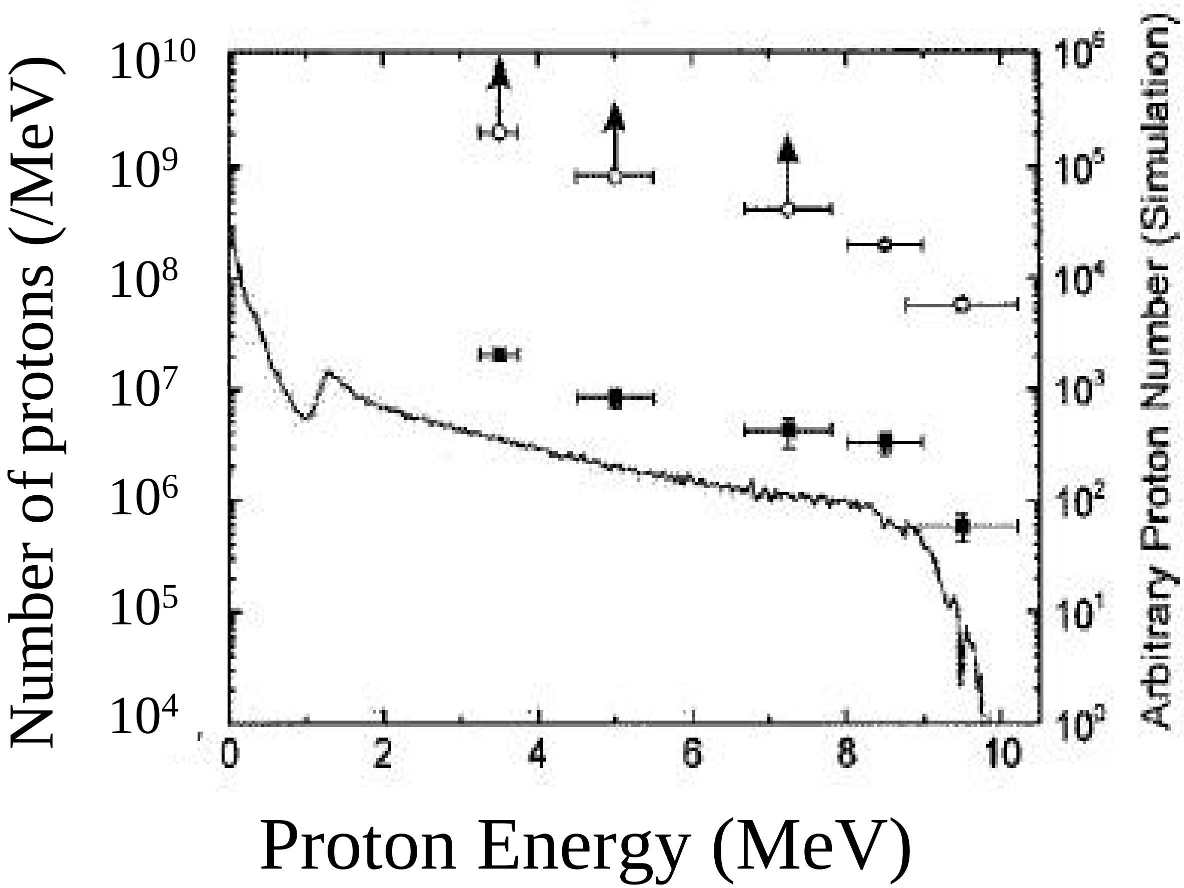
Neutrons



Unique source able to produce e^-, p, ions, neutrons, γ ?

Acceleration: $T_{eV} \propto E \times L$; laser: $E\Uparrow$ $L\Downarrow$ proximité source cible

Protons



S. Fritzler, V. Malka, G. Grillon, J. P. Rousseau, F. Burgy, E. Lefebvre, E. d'Humières, P. McKenna and K. W. D. Ledingham Appl. Phys. Lett 83, 3039 (2003)