



PALS in the area between high-energy density physics and high-energy chemistry

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Outline

- (a) decomposition of molecular and covalent solids exposed to intense XUV/X-ray radiation**
- (b) chemical consequences of large laser sparks in gases**



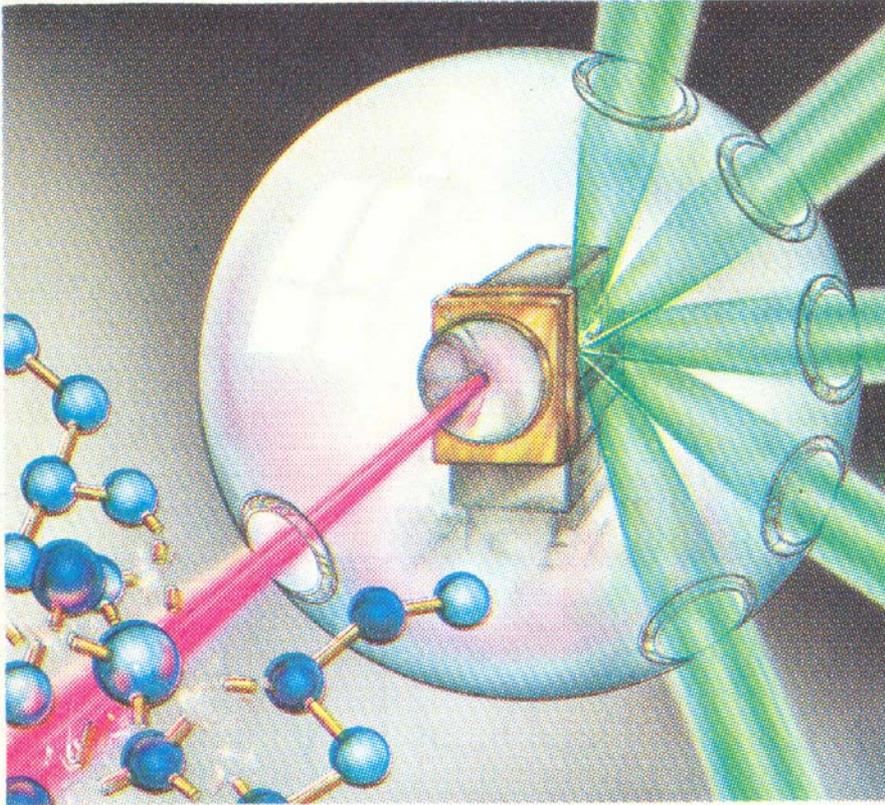
a) x-ray laser induced decomposition of solids

Why interaction of intense short-wavelength radiation with solid surfaces should be investigated?



1. diffraction-limited **nanstructuring** of solid surfaces for fabrication of **microelectronic and micromechanical elements** and devices
2. estimating and minimizing **damage to surfaces of heavily loaded XUV/x-ray optical elements** developed and used for guiding and focusing of short-wavelength laser beams
3. **durability assessments of materials** suggested for the **first walls of ICF reactors** and optical elements exposed to intense XUV/x-ray radiation in a laser-plasma interaction chamber
4. **determination of radiation field characteristics**: imaging of spatial energy distribution in a focused beam ablatively imprinted on the irradiated material and determination of pulse energy content

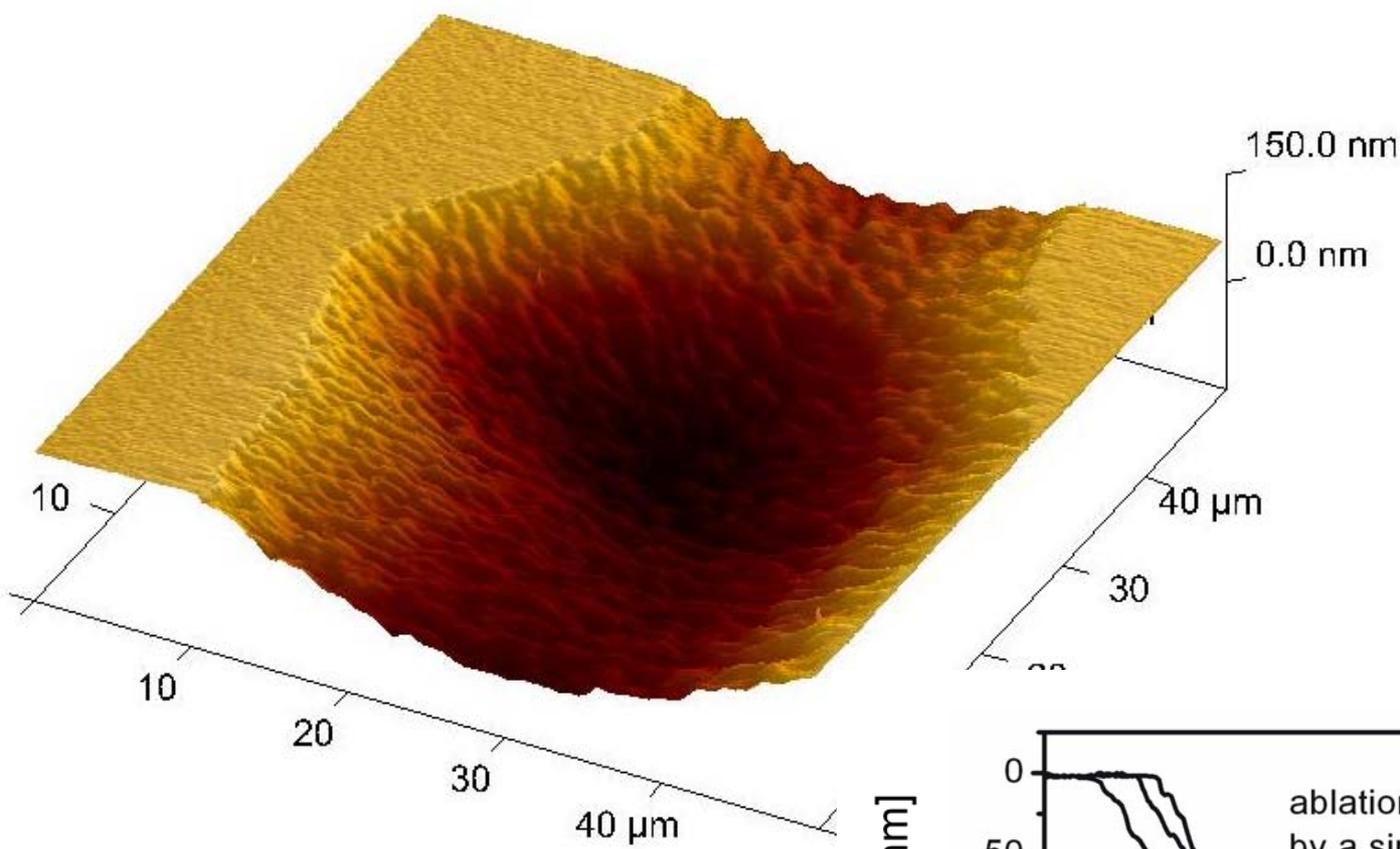
(Dessin de Fernando Cunha)



Casser des molécules, une des éventuelles applications du laser X. (p. 16)

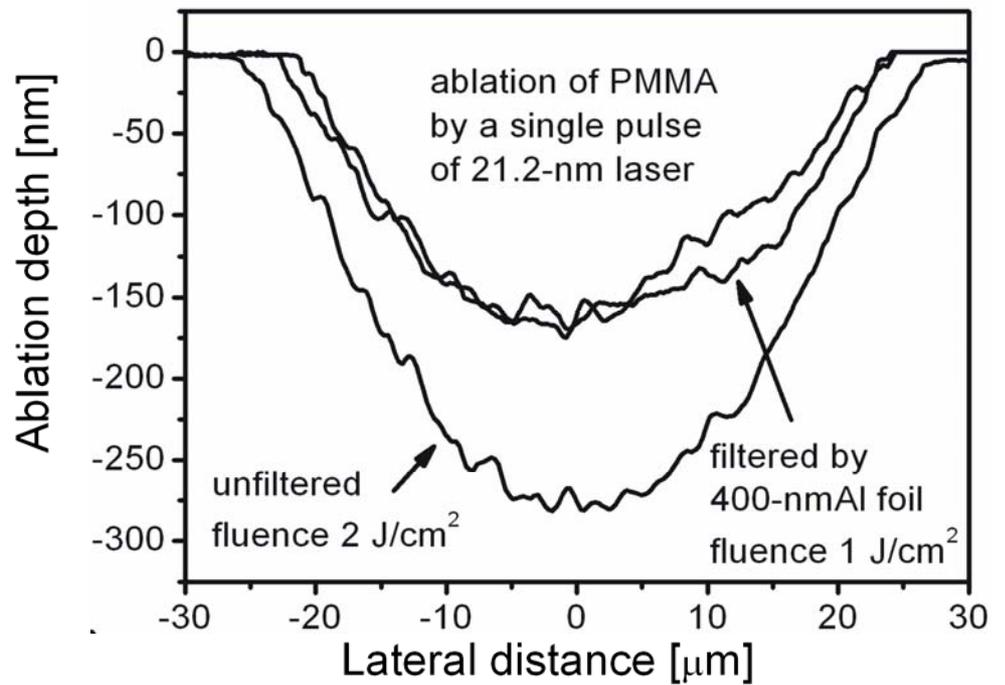
P. Jaeglé: Le laser à rayons X, *La Recherche* (184), 16-25 (Jan. 1987).

Decomposition of molecules, one of possible applications of X-ray lasers.



AFM image of crater ablated in PMMA by a focused beam of 21.2-nm laser radiation.

J. Microlith. Microfab. Microsyst.
4(3), 033007 (2005)

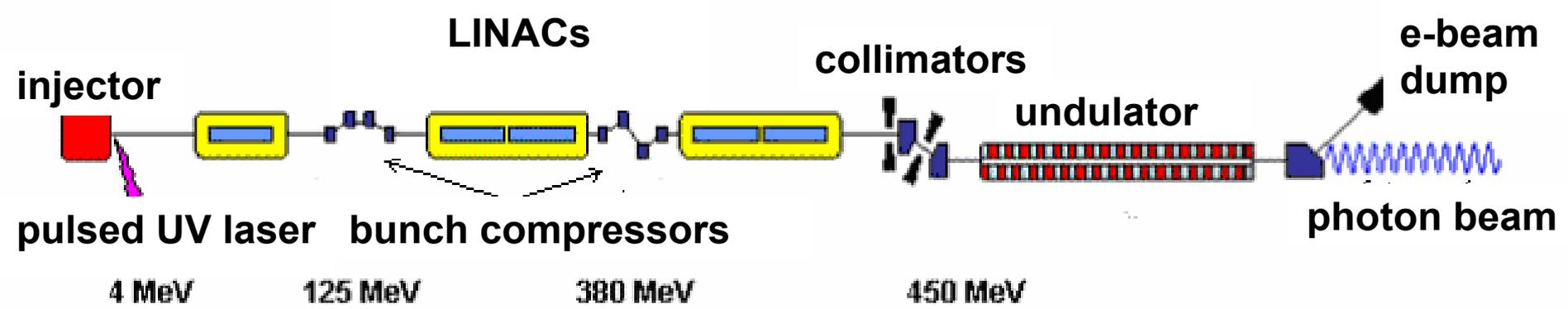


XUV/X-ray laser-decomposition of solids can be at least of two kinds

- 1) low fluence – multiple shots: **single photon (nonthermal) damage** [laser desorption]
- 2) high fluence – single shot: **collective (heat influenced) damage** [laser ablation]

D. A. G. Deacon: Optical coating damage and performance requirements in free electron laser, *Nucl. Instrum. Meth. Phys. Res.* **A250** (1-2), 283-288 (1986).

- 3) scientifically very interesting are intermediate irradiation conditions, i. e. few-shot near-threshold irradiation



FLASH (Free-electron LASer in Hamburg) – earlier VUV FEL, at the beginning TTF2 FEL

$\lambda = 32 \text{ nm}$ (tunable from **6 nm** to **60 nm**)

$E_{\text{sample}} = \sim 10 \text{ } \mu\text{J}$ ($\sim 10^{12}$ photons)

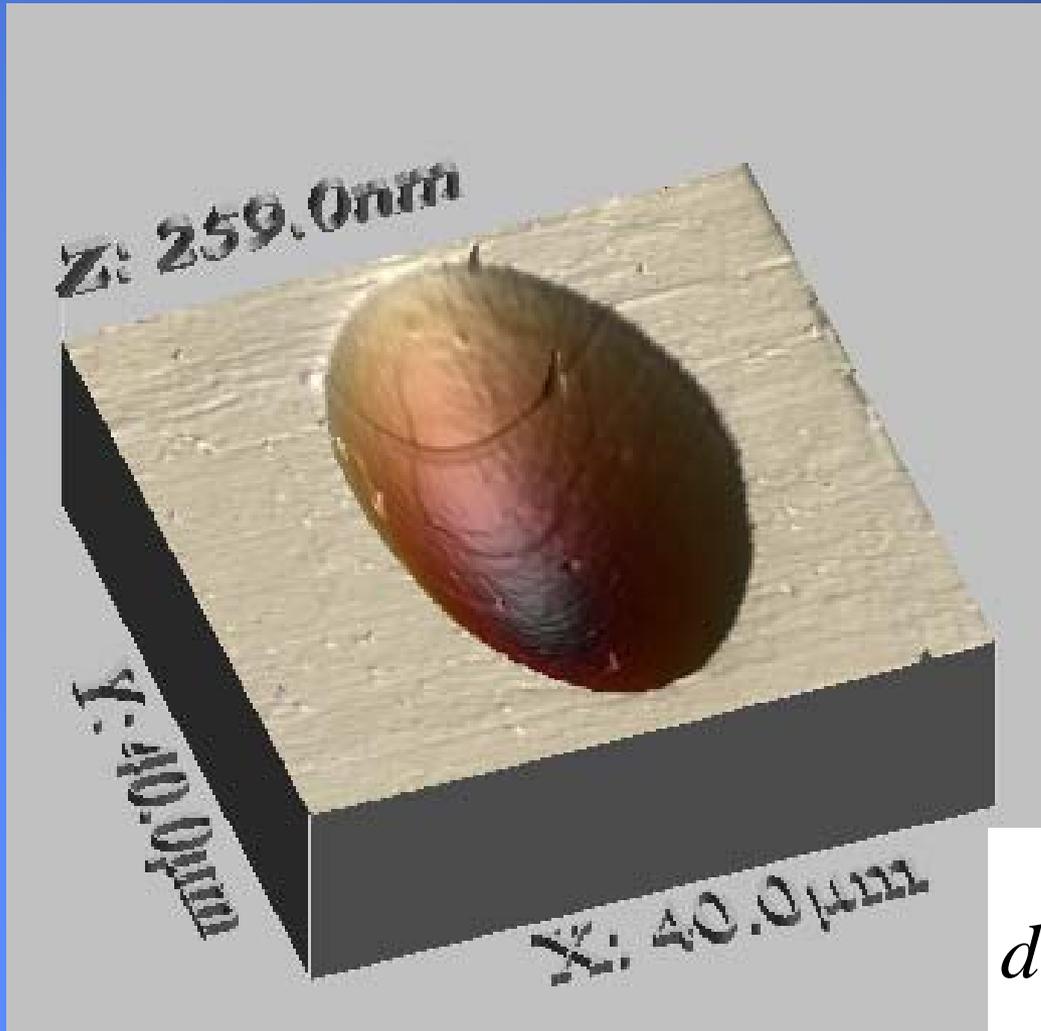
$t_p = 20 \text{ fs}$

$rr = 5 \text{ Hz}$



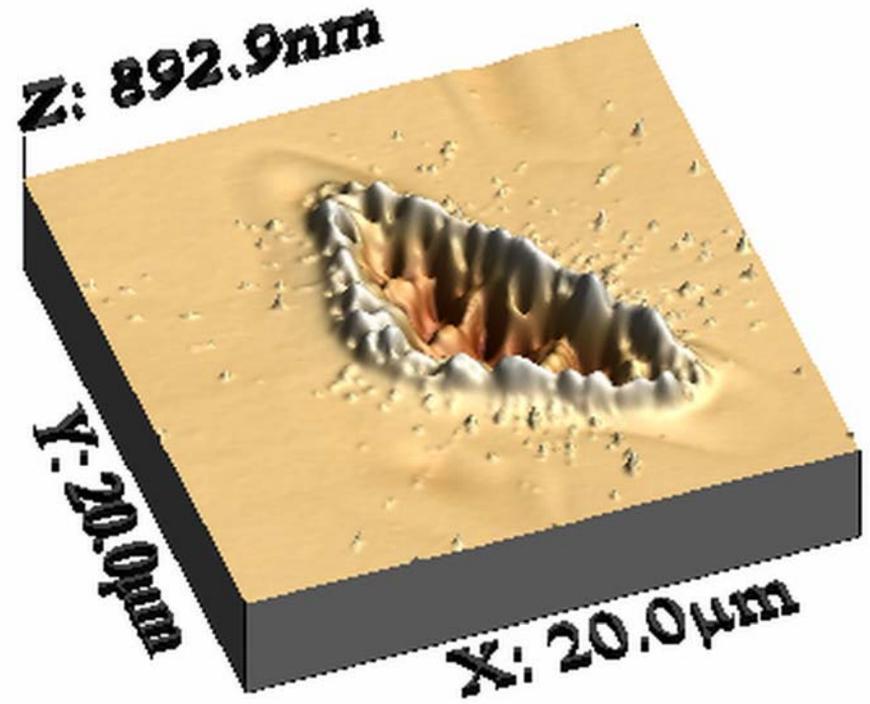
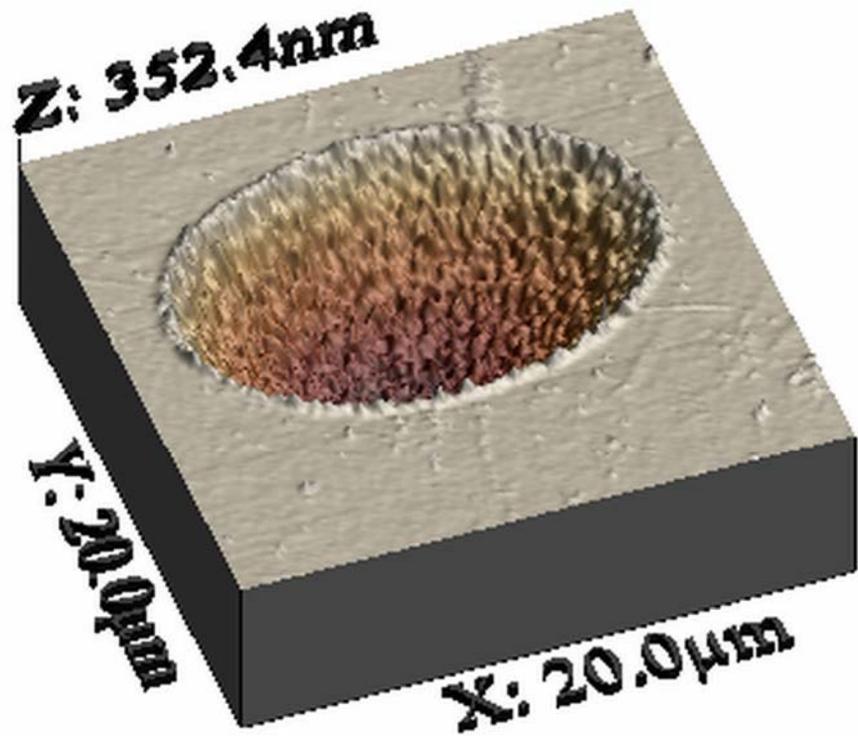
beam characteristics from shape of ablation crater – PMMA irradiated by FLASH at 21.7 nm

J. Chalupský et al.: *Opt. Express* **15**, 6036 (2007)



The crater shape is expected and found to be paraboloidal; it follows from the Gaussian beam profile {PMMA attenuation length l_{at} , pulse energy E , ablation threshold energy E_{th} , radial polar coordinate r , focal spot radius ρ }

$$d(r) = l_{at} \left(\ln \left(\frac{E_{pulse}}{E_{th}} \right) - \frac{r^2}{\rho^2} \right)$$



FLASH

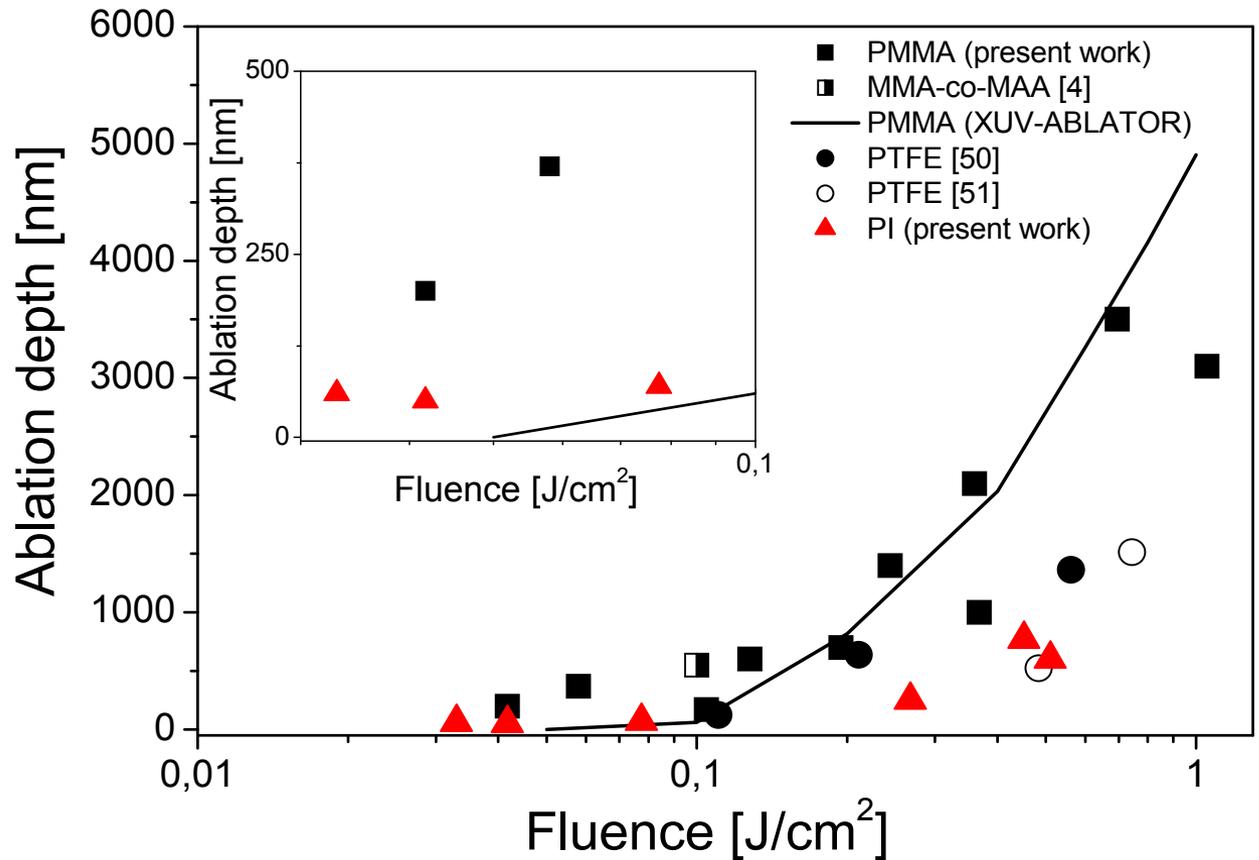
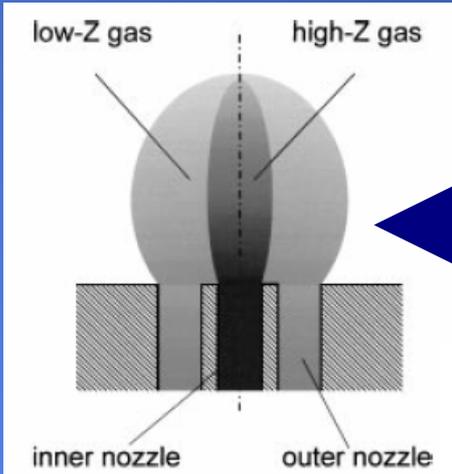
13.5 nm

7.0 nm

wavelength/pulse duration effects on ablation threshold in PMMA :

Laser wavelength (nm)	Pulse duration (fs)	Ablation threshold (mJ/cm ²)
32.5	15	1-2
21.7 / 21.2	10	11
	80000	300-500
13.5	10	30
7.0	10	56

PALS beam
E = 500 J
 $\lambda = 1315 \text{ nm}$



*Journal of
Microlith.
Microfab.
Microsyst.*
4, 033007
(2005)

Linac Coherent Light Source at SLAC

X-FEL based on last 1-km of existing linac

1.5-15 Å

Injector
at 2-km point

Existing 1/3 Linac (1 km)
(with modifications)

New e^- Transfer Line (340 m)

X-ray
Transport
Line (200 m)

Undulator (130 m)

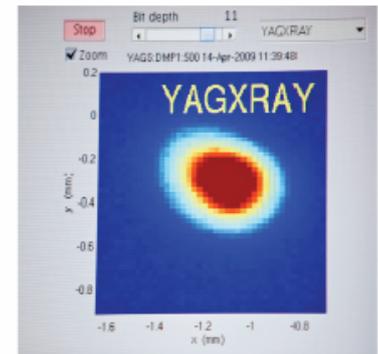
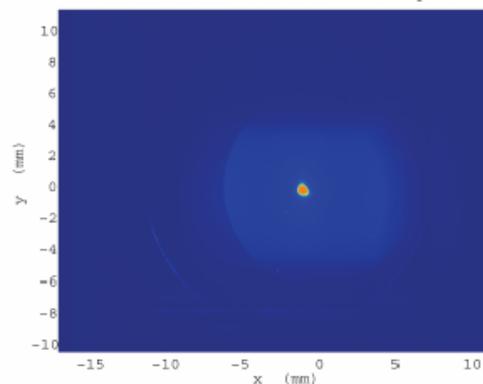
Near Experiment Hall
(underground)

Far Experiment
Hall (underground)

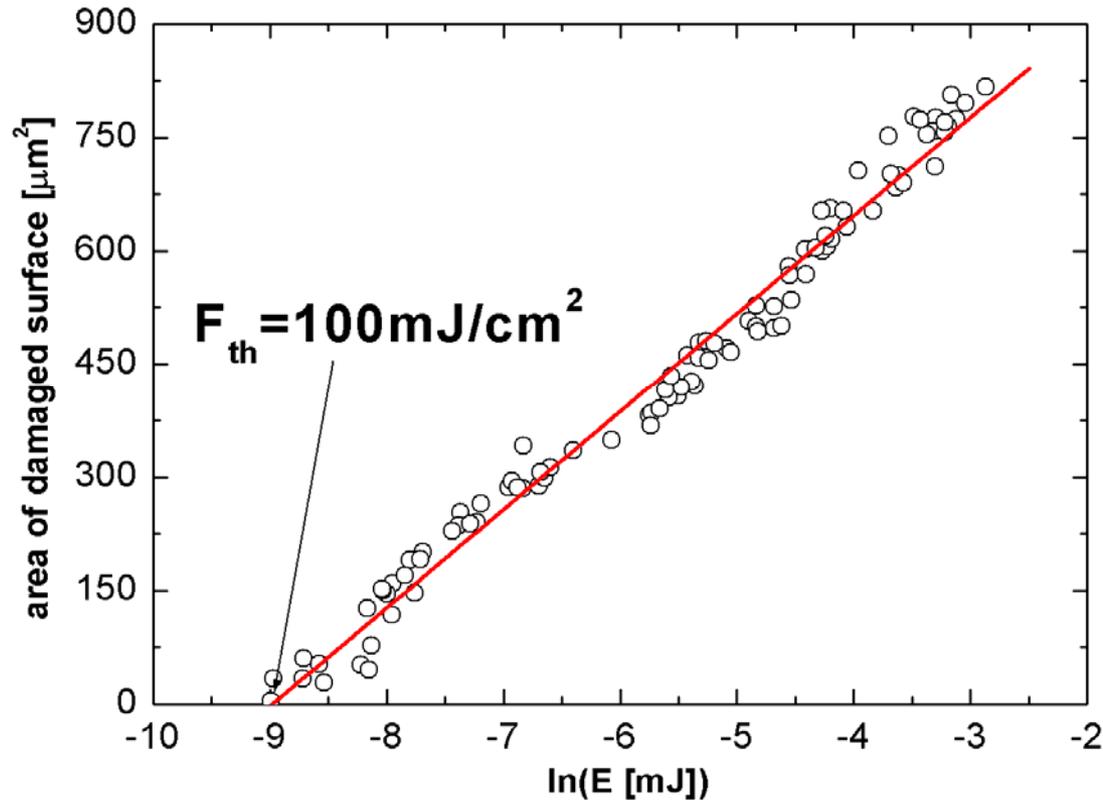


4-10-09 at LCLS: First FEL light at 1.5 Å

Profile Monitor YAGS:DMP1:500 11-Apr-2009 0



commissioning beamtimes
9/2009 (AMO) and 5/2010
(SXR): IOP-Prague+LBL+
LLNL+SLAC



Area of PMMA surface damaged by a single shot of 1.5-nm (830 eV) laser radiation vs. pulse energy. It shows clearly that ablation of PMMA by a single shot begins at $F_{th} = 100 \text{ mJ/cm}^2$. Wavelength scaling of FLASH results gives a value of $F_{th} = 420 \text{ mJ/cm}^2$. Very similar findings we got for inorganic dielectrics, e.g., PbWO_4 begins to ablate at 140 mJ/cm^2 of 1.5-nm radiation not at 400 mJ/cm^2 as estimated according to FLASH results.

conclusion (I)

We are studying an interaction of XUV/x-ray radiation with matter under the following conditions

from 100 nm to a few Å

from 10 fs to 10 ns

from 10^6 W/cm² to 10^{17} W/cm²

from 0° to 90°



b) laser-plasma chemistry

collaborating groups



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motivation



A few years after the laser's invention it was found that focusing a pulsed laser beam into a gas causes its dielectric breakdown in a part of the converging beam. A relatively hot, dense plasma is formed that is well defined in time and space. The process is called laser-induced dielectric breakdown (LIDB) while the phenomenon is called a laser spark.

Although the physical nature of laser sparks has been subject of numerous overviews, their chemical consequences surprisingly have not yet been discussed at such a level.

chemistry of laser sparks



**Inorganic and organic chemistry of laser sparks:
reviewing chemical reactions initiated by laser
plasmas**

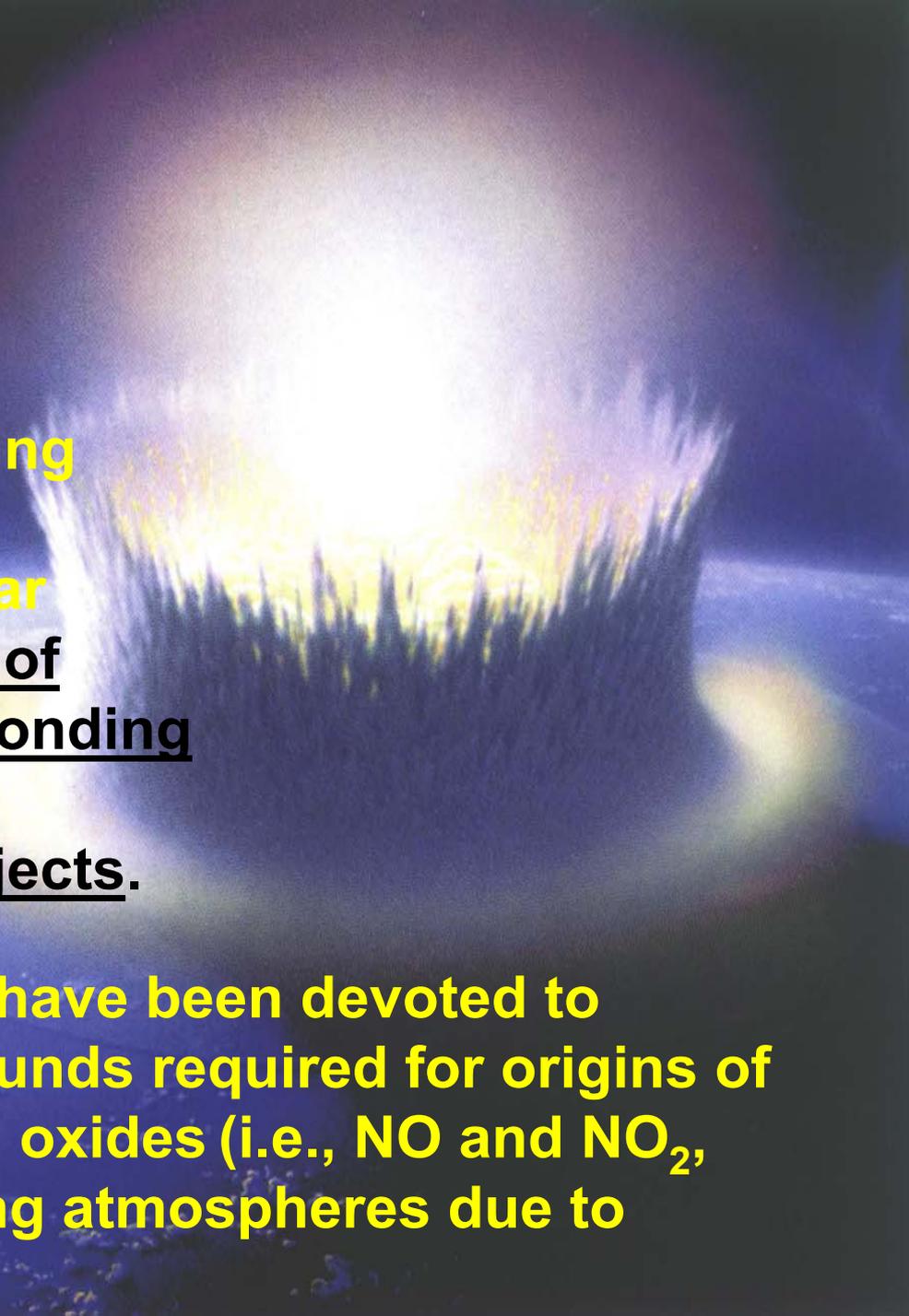
**Cosmogeochemistry with laser sparks:
laboratory simulation of high-energy density
events in planetary atmospheres**

**Physical chemistry / chemical physics of laser sparks:
What are the processes responsible for
laser-plasma induced chemical reactions?**

cosmogeoechemistry with laser sparks

The current research in laser-plasma chemistry received a strong, accelerating impulse from work utilizing LIDB in mixtures of molecular gases as laboratory models of planetary atmospheres responding to lightning or high-velocity impact of extraterrestrial objects.

Extensive chemical studies have been devoted to synthesis of organic compounds required for origins of life and to generate nitrogen oxides (i.e., NO and NO₂, formed in nitrogen containing atmospheres due to lightning).



What are the processes responsible for laser-plasma induced chemical reactions?



The laser pulse energy deposited in the LIDB plasma penetrates into the surrounding gas through:

- 1) short-wavelength radiation (i.e. UV-VUV, XUV, x-ray) and energetic particles emitted from the plasma core,
- 2) a shock wave created by LIDB, and
- 3) mixing and interaction of the plasma containing huge amounts of highly reactive species (atoms, atomic and molecular ions, free radicals); thermal waves propagating through the gas.

The role of these processes play within real LIDB plasmas are determined by the laser pulse characteristics controlling both the electron temperature and other plasma parameters and their ability to initiate particular chemical reactions in a given molecular system.

OUR MISSION IN THE FIELD

instead of many small laser sparks produced
by a high-repetition laser

we are dealing with
a single, large laser spark

WHY LARGE LASER SPARKS?

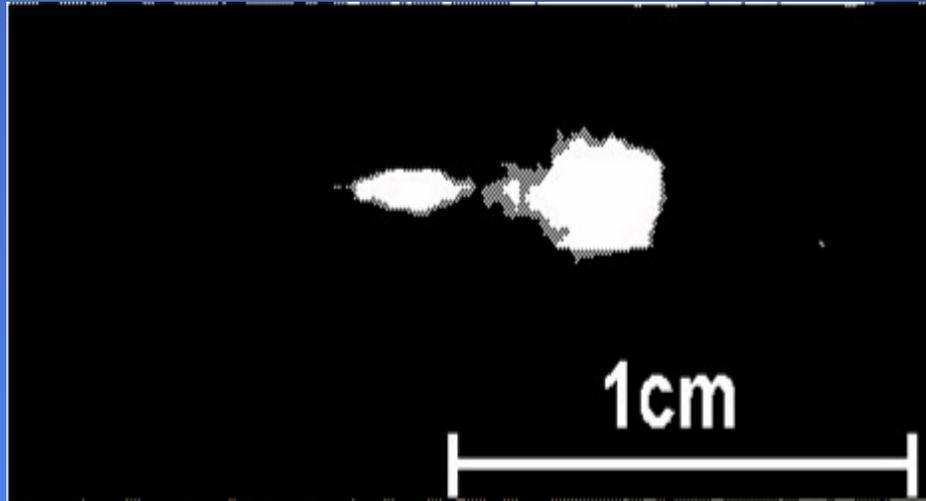


How to produce them? – using the high power laser system, e.g., PALS

their advantages over small, high-repetition lasers:

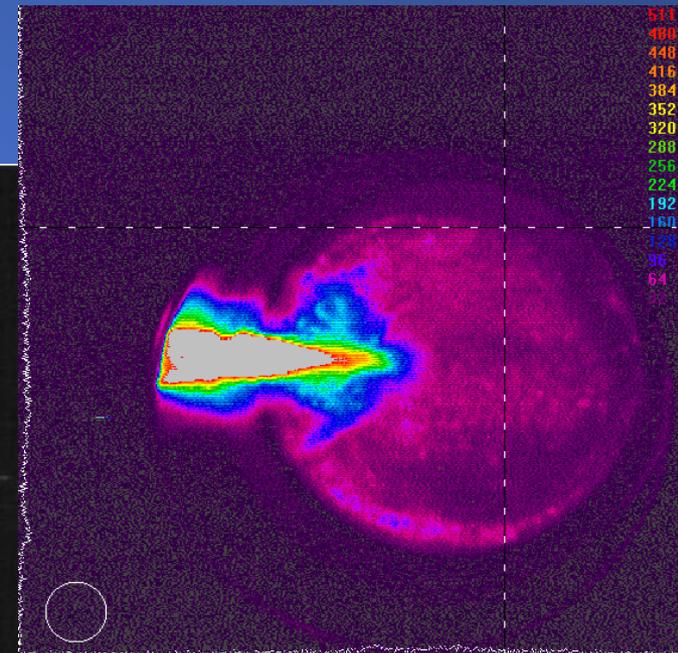
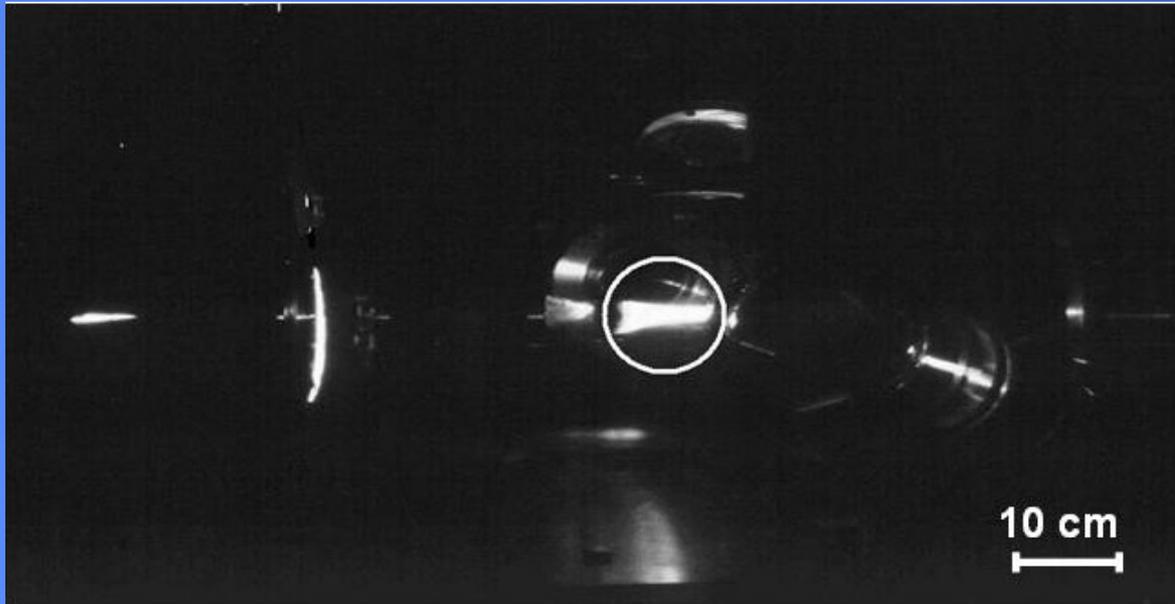
- higher pulse energy, shorter pulse duration \Rightarrow higher intensity \Rightarrow much higher volumes of the LIDB plasma
- greater amount of generated products
- more realistic laboratory simulation of a single high-energy-density atmospheric event
- a single pulse eliminates the influence of LIDB plasma on products formed earlier

high-repetition, low-power laser \Rightarrow laser spark



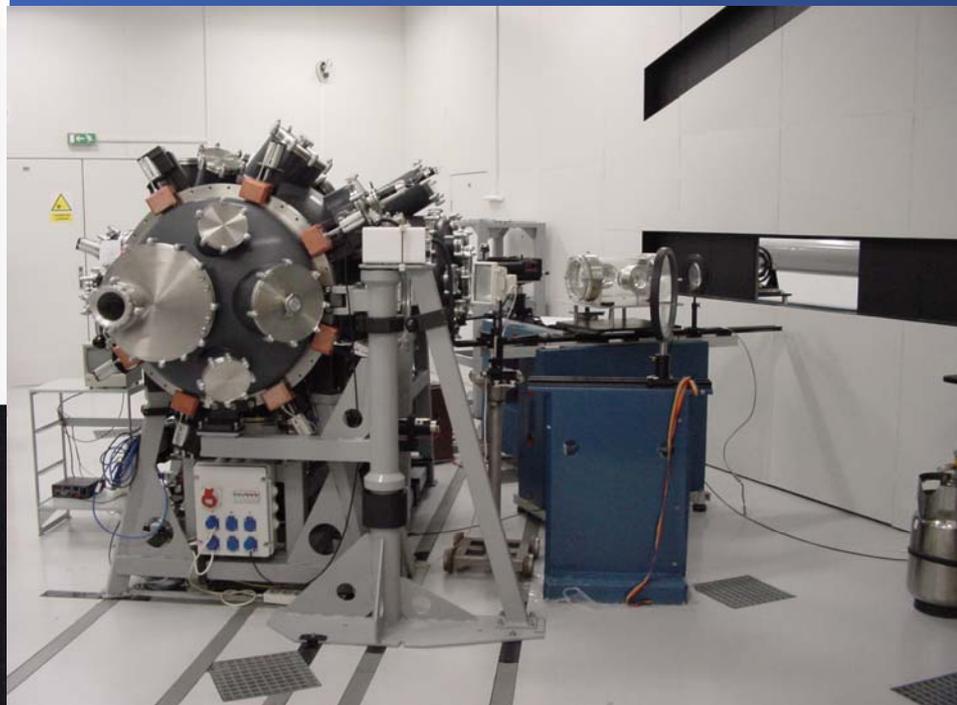
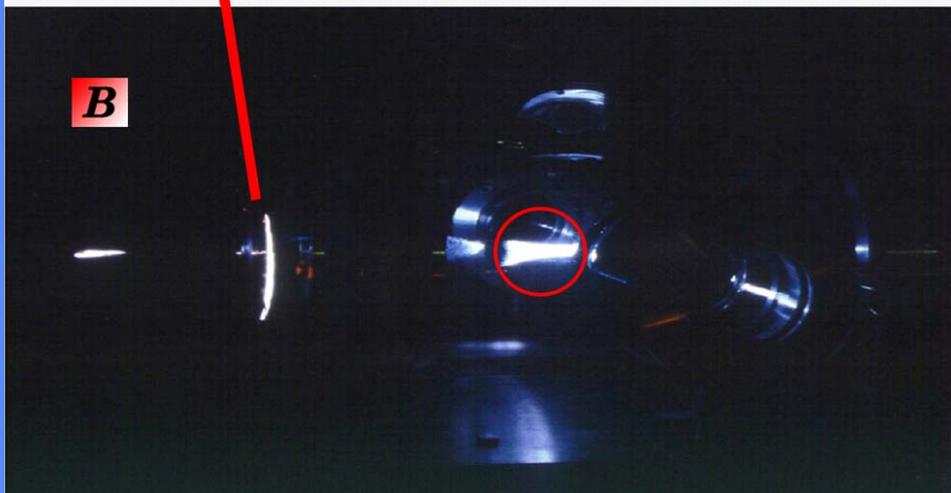
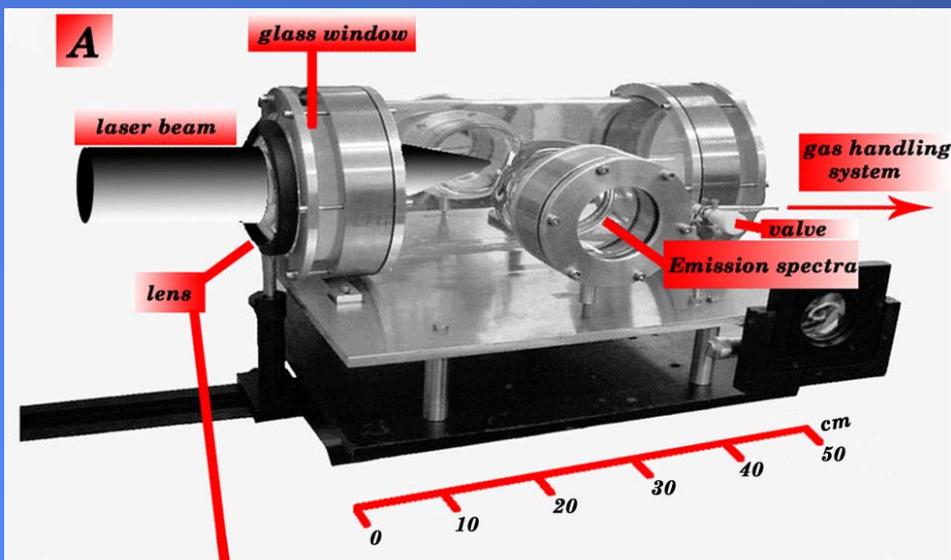
3-J TEA CO_2 laser, air; by courtesy of Prof. Vrbová

high-power laser \Rightarrow large laser spark



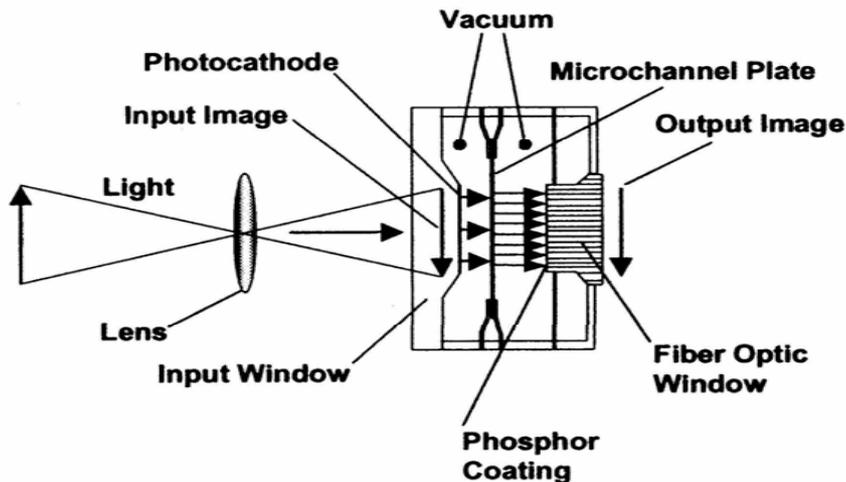
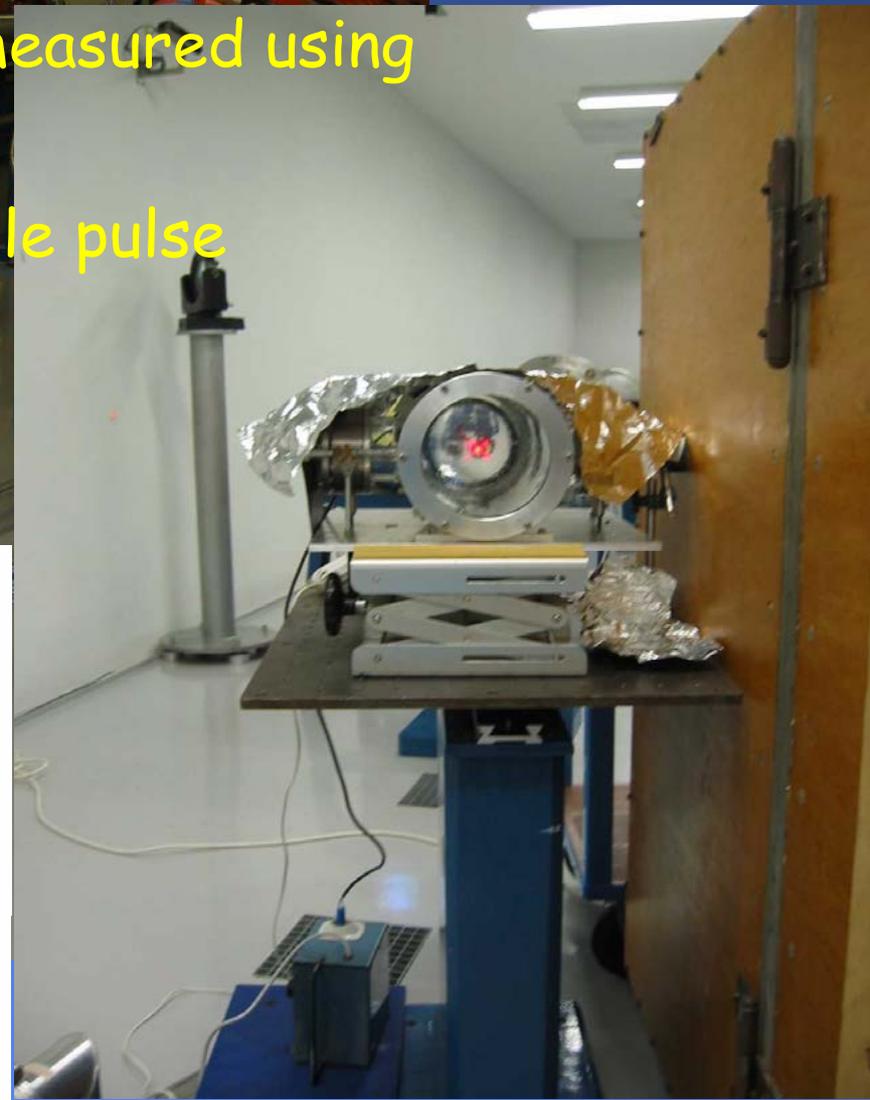
0.5-kJ iodine laser (PALS)

the static LIDB experiment, 15-L gas cell (a)



the static LIDB experiment, 15-L gas cell (b)

- optical emission spectra were measured using spectrometer with ICCD head
- spectra were collected in a single pulse
- spectral range: 350-1000 nm
- laser pulse energy: 100-850 J



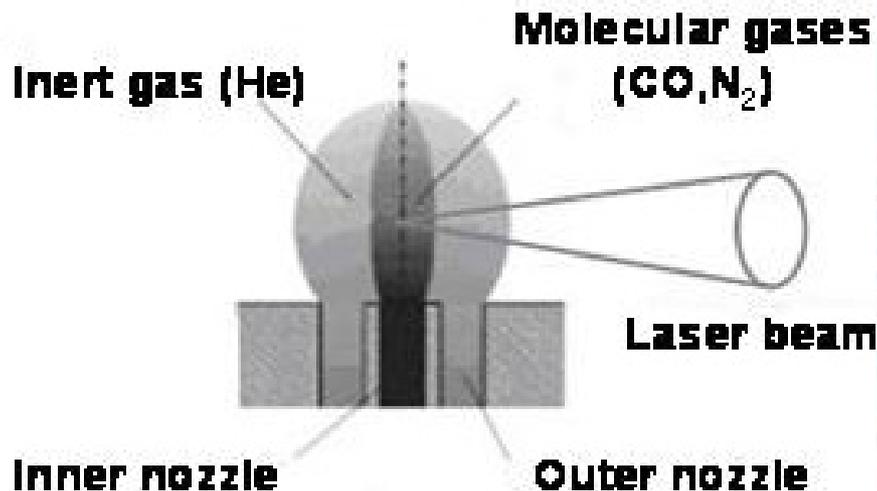
molecular emission bands dominate all the emission spectra in static arrangement

- 1) **CO-N₂-H₂O**: C₂ radicals (Swan and Deslandres-D'Azambuja system); C₃ radical (Swings system) and CN radical (violet system).
- 2) **CO₂-N₂-H₂O**: only bands belonging to the CN radical (violet system) were found
- 3) **NH₃-CH₄-H₂-H₂O**: C₂ radical (Swan system) predominated in the, with less intense bands of the CN radical (violet system).

The identified molecular bands were also used for determination of plasma parameters.

the dynamic LIDB experiment, gas-puff target (MUT, Warsaw)

- allows investigation of the hot core of the LIDB plasma (soft x-ray radiation, highly charged particles)
- allows us to compare optical spectra when the hot core is not surrounded with cold gas
- all spectra were measured in one pulse
- laser pulse energy varied from 100 J to 600 J

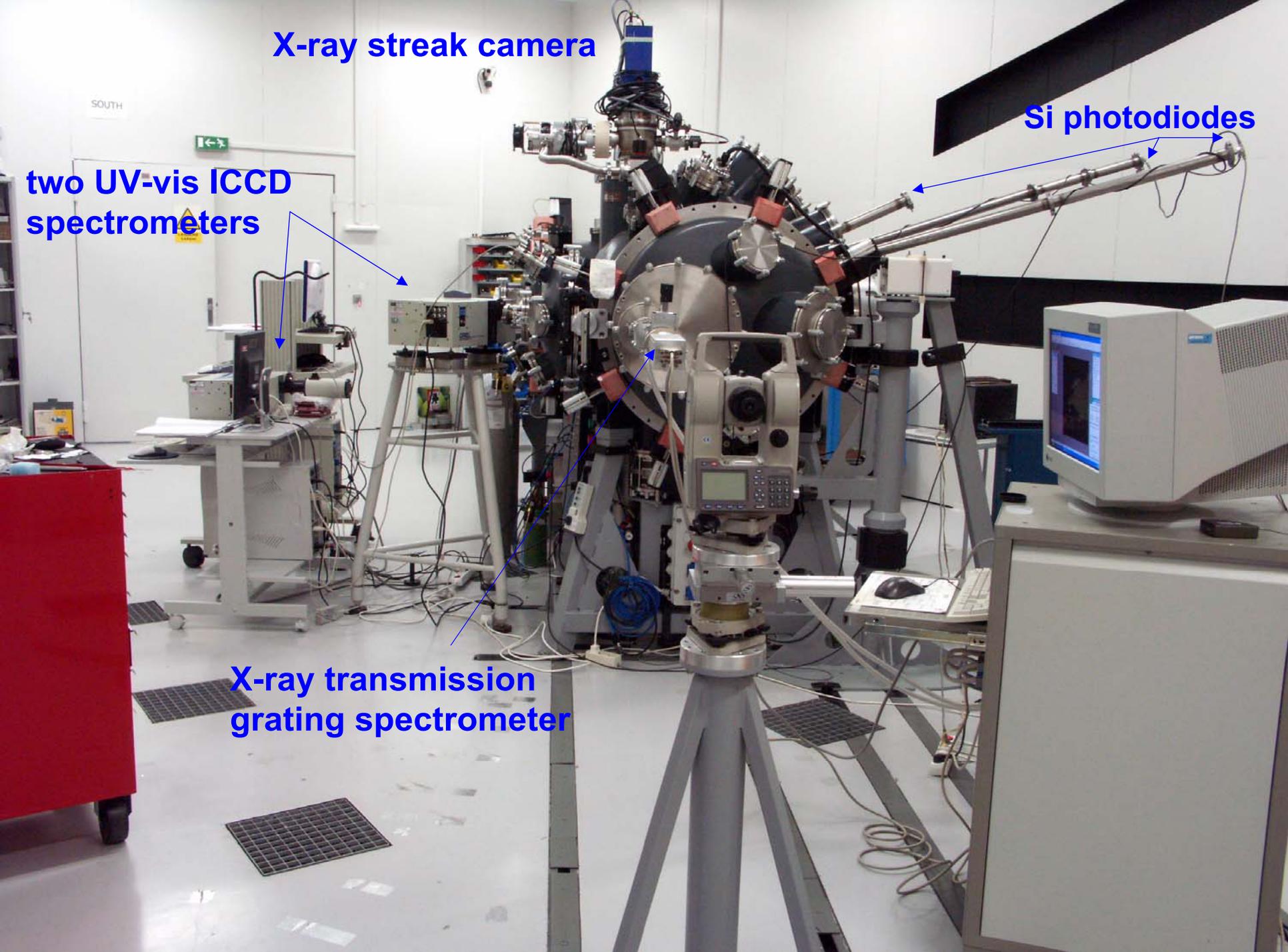


X-ray streak camera

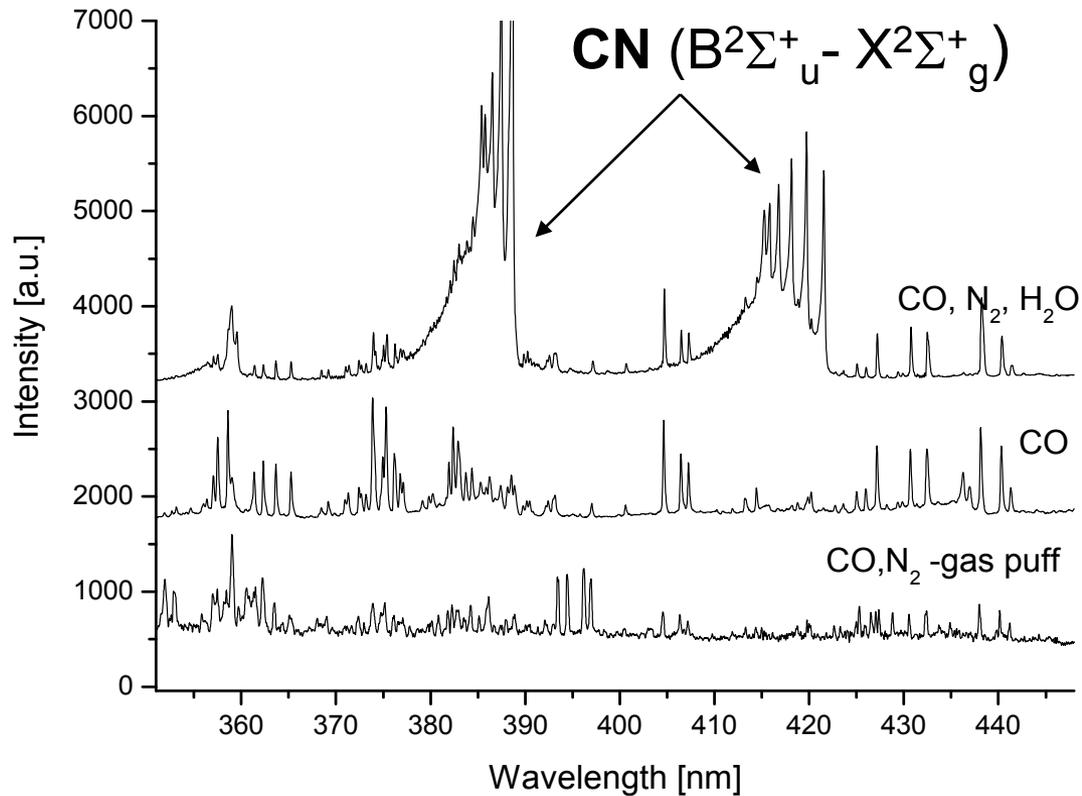
Si photodiodes

two UV-vis ICCD spectrometers

X-ray transmission grating spectrometer



UV-vis emission spectra



no molecular bands were identified

the presence of the cold molecular gas surrounding the hot core of the LIDB plasma is a necessary condition for formation of complex molecular species

conclusion (II)



The study of the chemical consequences of LIDB in molecular gases is a well established, although not fully developed, branch of laser chemistry.

A laser spark acts on surrounding gas both thermally (by the expanding plasmas thermal and shock waves) and photochemically (mainly by short-wavelength electromagnetic radiation, i.e., UV, VUV, XUV, and x-rays). Although some results related to individual mechanisms may be found in the laser-plasma chemistry literature, there are no publications dealing in detail with all factors and providing a complex picture of the chemical consequences of LIDB in molecular gases.

**THANK YOU
FOR YOUR ATTENTION**