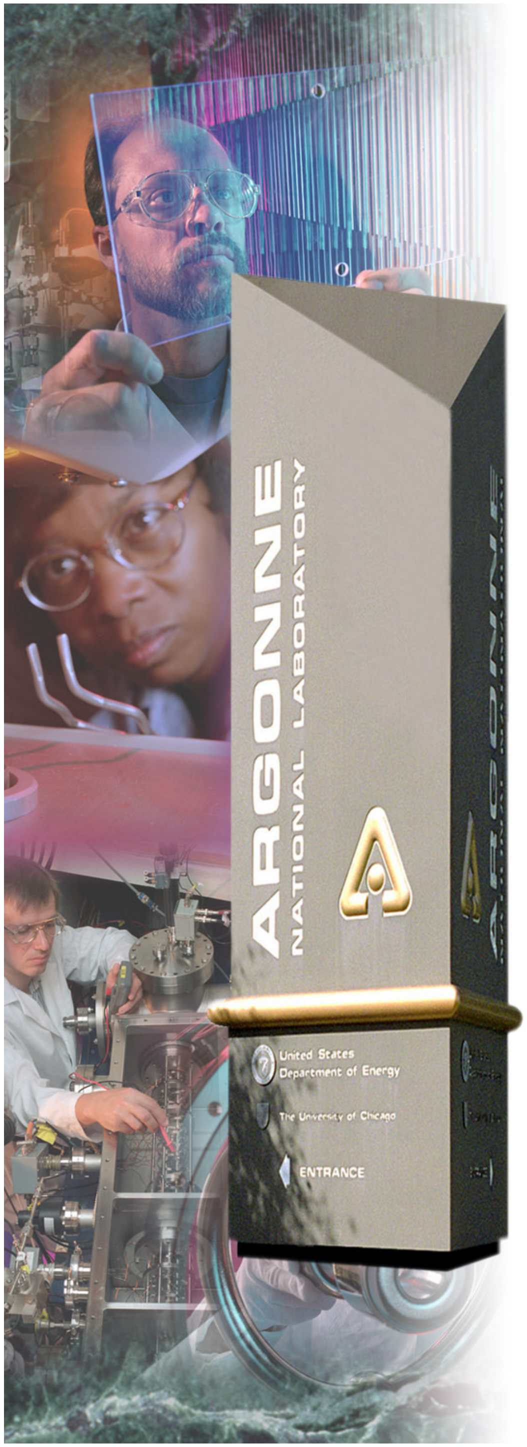

Ultrafast Radiation Chemistry and the Development of Laser Based Electron Sources*

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International Conference on Transient Chemical Structures in Dense Media, Paris, France
March 14, 2005

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Outline

- **Radiation Chemistry**
- **Photochemistry Studies**
- **Development of ultrafast electron source**

Radiation Chemistry

The interaction between ionizing radiation and molecules/atoms is the leading cause of chemical reactions in our universe

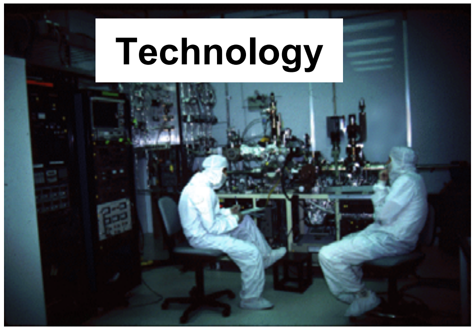
Here on Earth


Nuclear Energy


Radio Therapy

Waste/Environment

Radiation Chemistry


Technology

Condensed Phase Chemical Physics

Secondary Effects (yields etc.) known Cause?

Ultrafast Radiation Chemistry

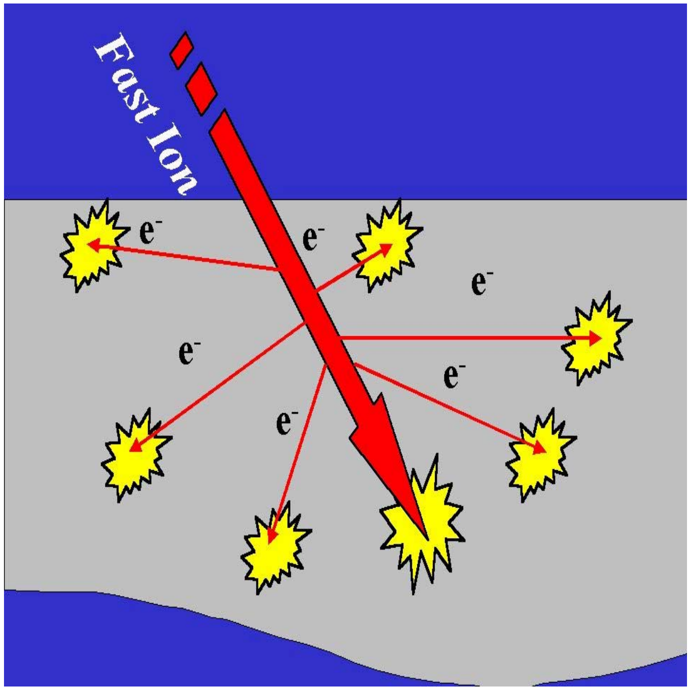
- **Understanding of high energy (radiation) chemistry in the condensed phase**

- **Primary (ultrafast) events are very important, determine secondary events**

- **Primary events in radiation chemistry virtually unknown, except for theoretical models**

- **We are developing a laser based subps source of ionizing radiation and x-rays**

The Spur – where it all begins



As ionizing radiation passes through condensed matter it produces secondary electrons (spurs) along its track. The secondary electrons are responsible for most of the resultant chemistry.

Several energetic and reactive species are produced in close proximity leading to complicated first and second order chemical reactions.

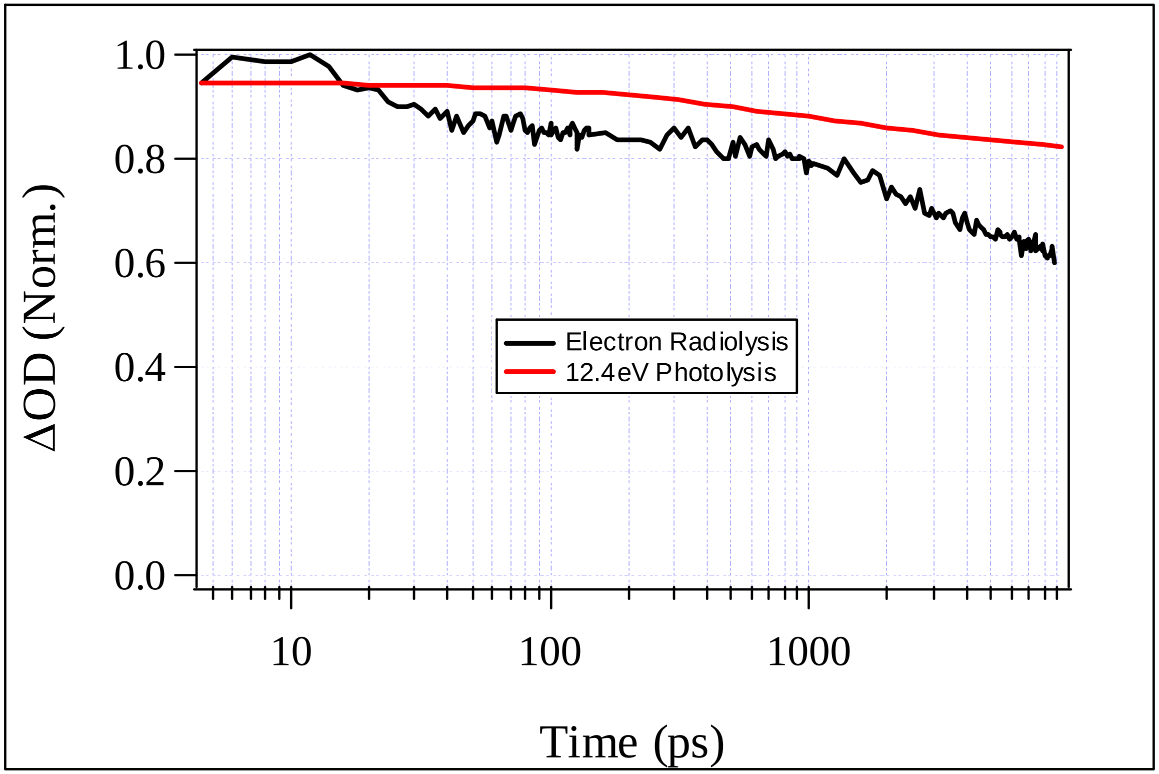
In water each spur contains ~2-3 secondary electrons. Spurs are ~100nm apart, 1-5nm diameter

The lack of selection rules combined with the high local concentration of energetic species and energy deposition makes radiolysis very different (much more complex) than photochemistry

Primary processes in radiation chemistry

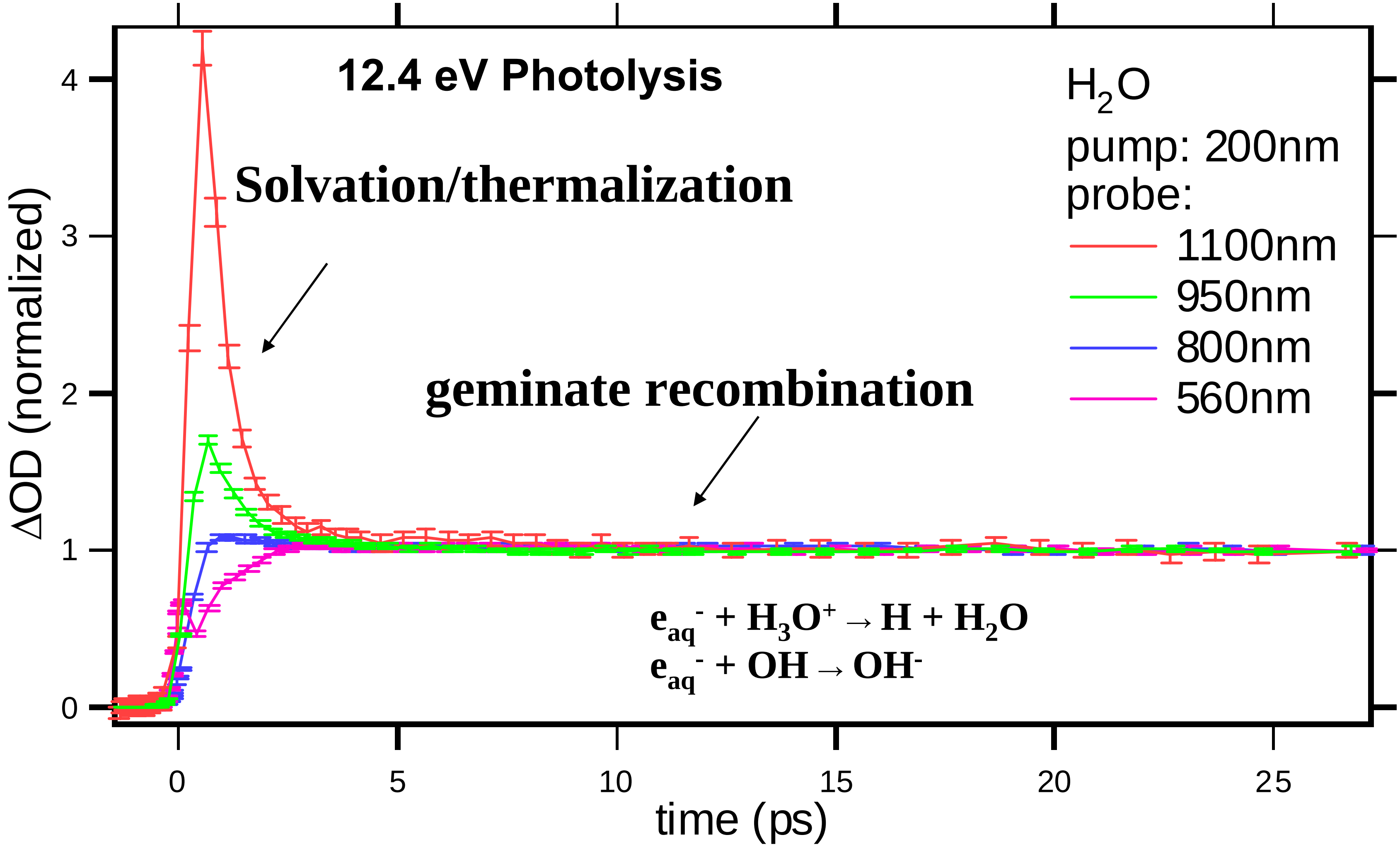
- Use a combination of ultrafast pulse radiolysis, x-ray, and ultrafast laser techniques to dissect the spur
- The most important primary processes that we wish to study are – energy deposition, thermalization, solvation, pre-thermalization chemistry, initial distribution of products
- We start with photoionization studies

Radiolysis vs Photolysis – e^-_{aq} in Water



- ❑Different electron recombination kinetics due to intra and inter-spur recombination
- ❑Photolysis isolated ionization events

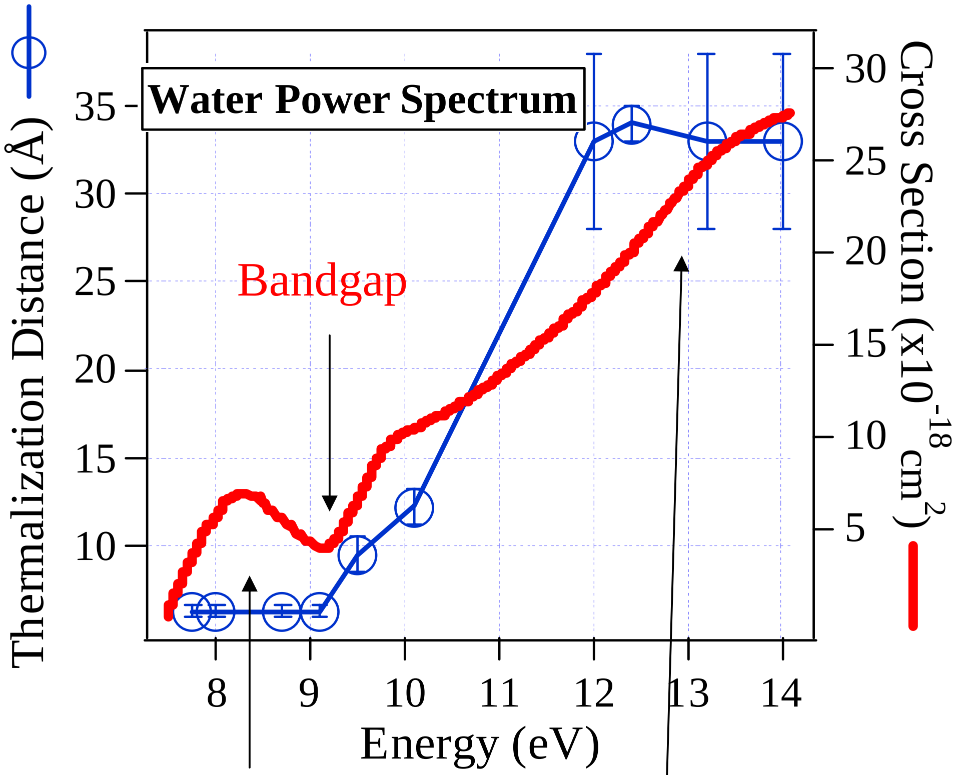
Fs Laser Spectroscopy – dynamics of photejected electrons



$e_{aq}^{-} + H_3O^{+} \rightarrow H + H_2O$

$e_{aq}^{-} + OH \rightarrow OH^{-}$

Water Photoionization



CTTS or Dissociation?

Conduction Band?

This is a very significant result

The Issues

-Mechanism(s)for electron production above and below the bandgap, electronic structure

-Spectroscopic identification of the various forms of e^-_{pre}

-Chemical reactivity of e^-_{pre} , which forms are more reactive? e.g., H_2 production

-Role of the solvent in electron thermalization/solvation

The Solution

Map out the spectral evolution of e^-_{aq} as a function of ionization energy

Probing the Liquid Conduction Band

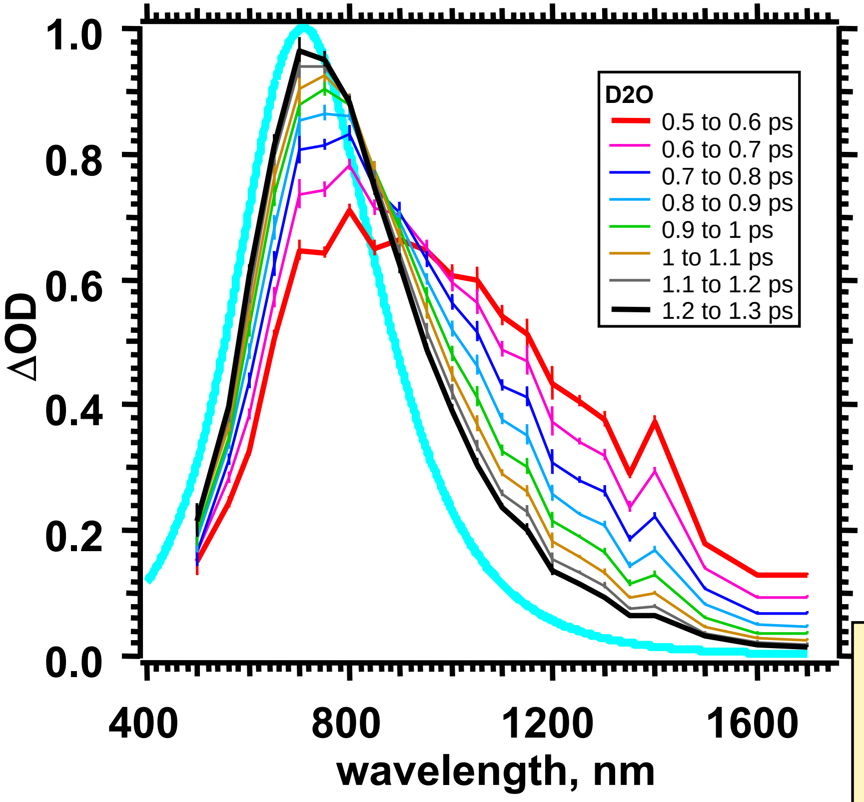
- **Spectral evolution (500nm-1700nm) of the electron spectrum following 2x6.2eV (12.4eV) ionization of H_2O and D_2O**

- **~3eV above the bandgap**

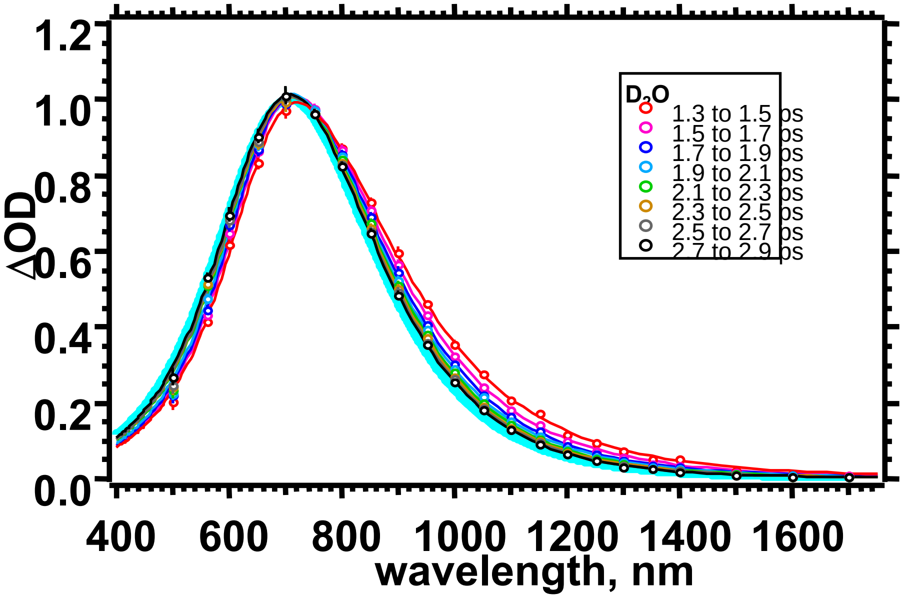
- **Geminate kinetics → σ ~ 25Å (similar to radiolysis ~35Å)**

12.4 eV (2x6.2eV) Water Photoionization

Solvation



Thermalization



Scheme

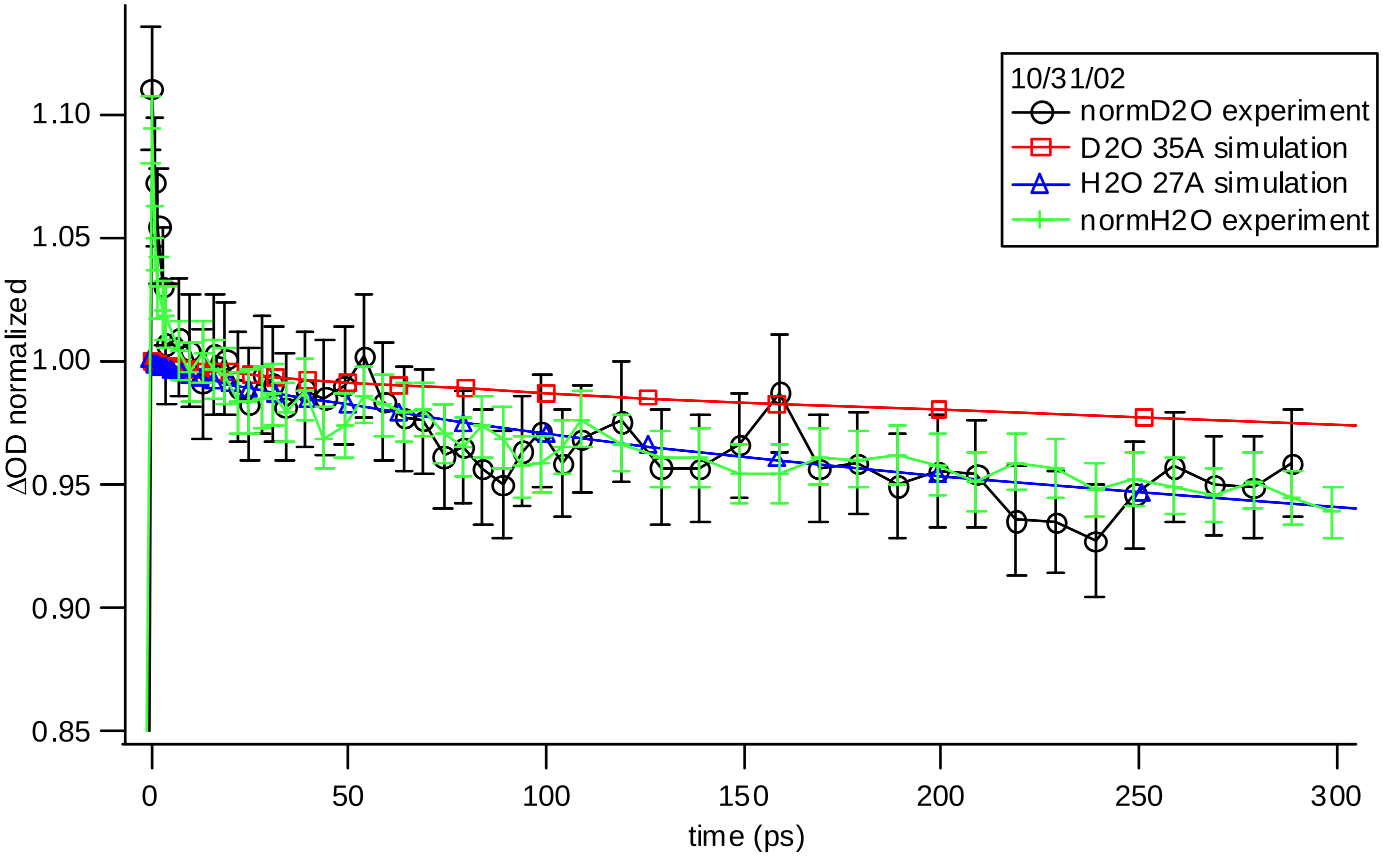
$e^-_{cb} \rightarrow\rightarrow$ $e^-_{NE}(hot) \rightarrow e^-_{aq}$

Nonradiative relaxation-> vibrations of solvent appear to play a role in both thermalization and solvation

Need theory

Geminate Kinetics 12.4eV Isotope Effect

- **H_2O vs D_2O**



Geminate Kinetics 12.4eV Isotope Effect

- No isotope on the shape of the geminate recombination kinetics

- Using independent pairs model

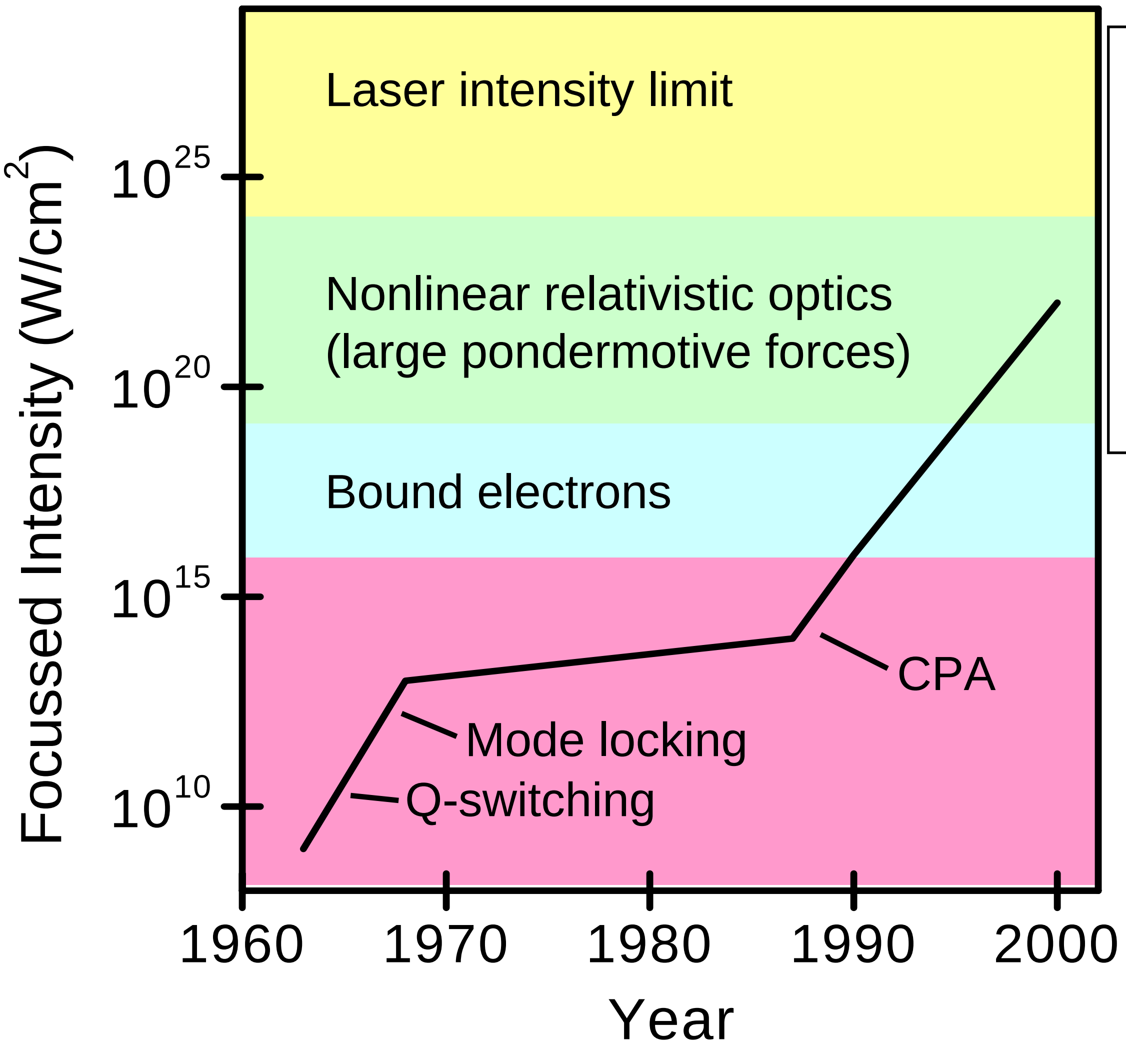
  escape distance for H_2O=2.4nm

  D_2O=2.1nm

Expect longer distance in heavy water because of smaller energy of accepting OD modes

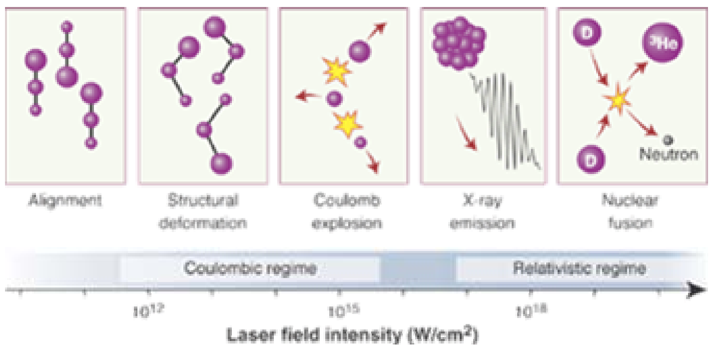
The narrower distribution in heavy water suggests that there is some competition between autoionization and direct ionization

Generation of Ultrahigh Peak Powers: Chirped Pulse Amplification

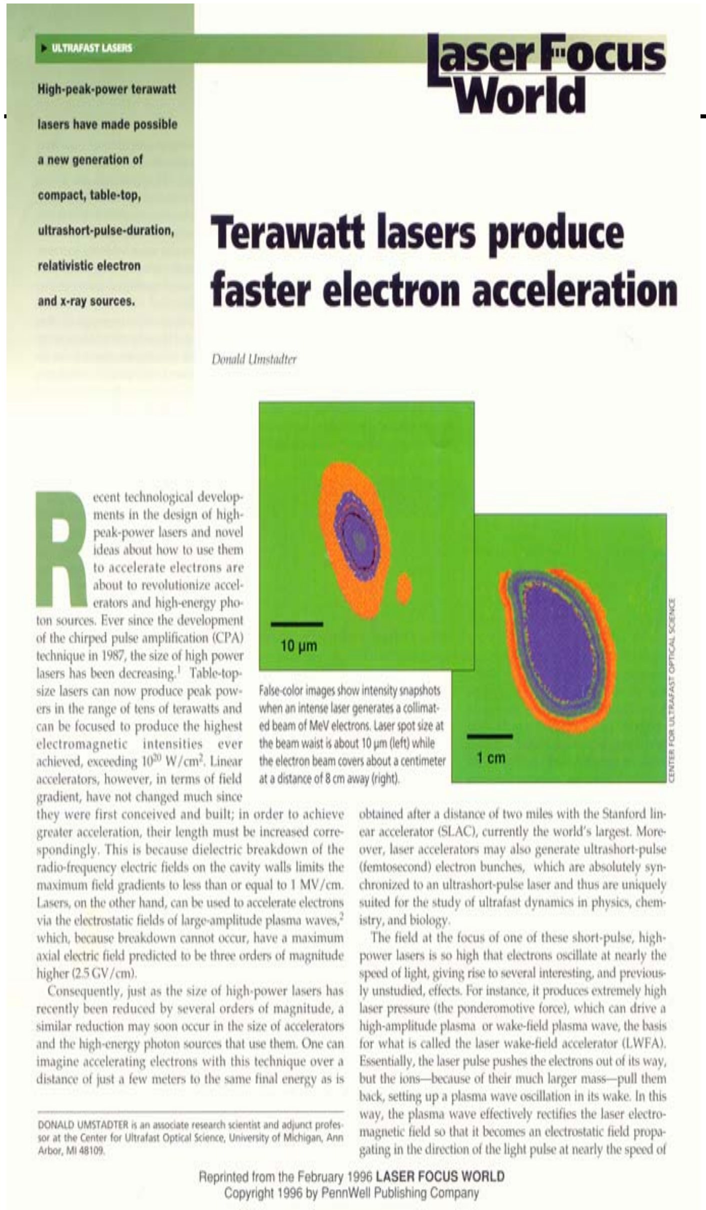


Recent advances in laser technology that have opened up new areas of research in physics and chemical physics

and radiation chemistry?



K. Yamanouchi *Science, 295 1659 (2002)*



ULTRAFAST LASERS

Laser Focus World

High-peak-power terawatt lasers have made possible a new generation of compact, table-top, ultrashort-pulse-duration, relativistic electron and x-ray sources.

Terawatt lasers produce faster electron acceleration

Donald Umstadter

False-color images show intensity snapshots when an intense laser generates a collimated beam of MeV electrons. Laser spot size at the beam waist is about 10 µm (left) while the electron beam covers about a centimeter at a distance of 8 cm away (right).

CENTER FOR ULTRAFAST OPTICAL SCIENCE

Recent technological developments in the design of high-peak-power lasers and novel ideas about how to use them to accelerate electrons are about to revolutionize accelerators and high-energy photon sources. Ever since the development of the chirped pulse amplification (CPA) technique in 1987, the size of high power lasers has been decreasing.[1] Table-top-size lasers can now produce peak powers in the range of tens of terawatts and can be focused to produce the highest electromagnetic intensities ever achieved, exceeding 10^{20} W/cm^2. Linear accelerators, however, in terms of field gradient, have not changed much since they were first conceived and built; in order to achieve greater acceleration, their length must be increased correspondingly. This is because dielectric breakdown of the radio-frequency electric fields on the cavity walls limits the maximum field gradients to less than or equal to 1 MV/cm. Lasers, on the other hand, can be used to accelerate electrons via the electrostatic fields of large-amplitude plasma waves,[2] which, because breakdown cannot occur, have a maximum axial electric field predicted to be three orders of magnitude higher (2.5 GV/cm).

Consequently, just as the size of high-power lasers has recently been reduced by several orders of magnitude, a similar reduction may soon occur in the size of accelerators and the high-energy photon sources that use them. One can imagine accelerating electrons with this technique over a distance of just a few meters to the same final energy as is obtained after a distance of two miles with the Stanford linear accelerator (SLAC), currently the world's largest. Moreover, laser accelerators may also generate ultrashort-pulse (femtosecond) electron bunches, which are absolutely synchronized to an ultrashort-pulse laser and thus are uniquely suited for the study of ultrafast dynamics in physics, chemistry, and biology.

The field at the focus of one of these short-pulse, high-power lasers is so high that electrons oscillate at nearly the speed of light, giving rise to several interesting, and previously unstudied, effects. For instance, it produces extremely high laser pressure (the ponderomotive force), which can drive a high-amplitude plasma or wake-field plasma wave, the basis for what is called the laser wake-field accelerator (LWFA). Essentially, the laser pulse pushes the electrons out of its way, but the ions—because of their much larger mass—pull them back, setting up a plasma wave oscillation in its wake. In this way, the plasma wave effectively rectifies the laser electromagnetic field so that it becomes an electrostatic field propagating in the direction of the light pulse at nearly the speed of

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Reprinted from the February 1996 **LASER FOCUS WORLD**

In the relativistic regime it becomes possible to generate subps e^- pulses

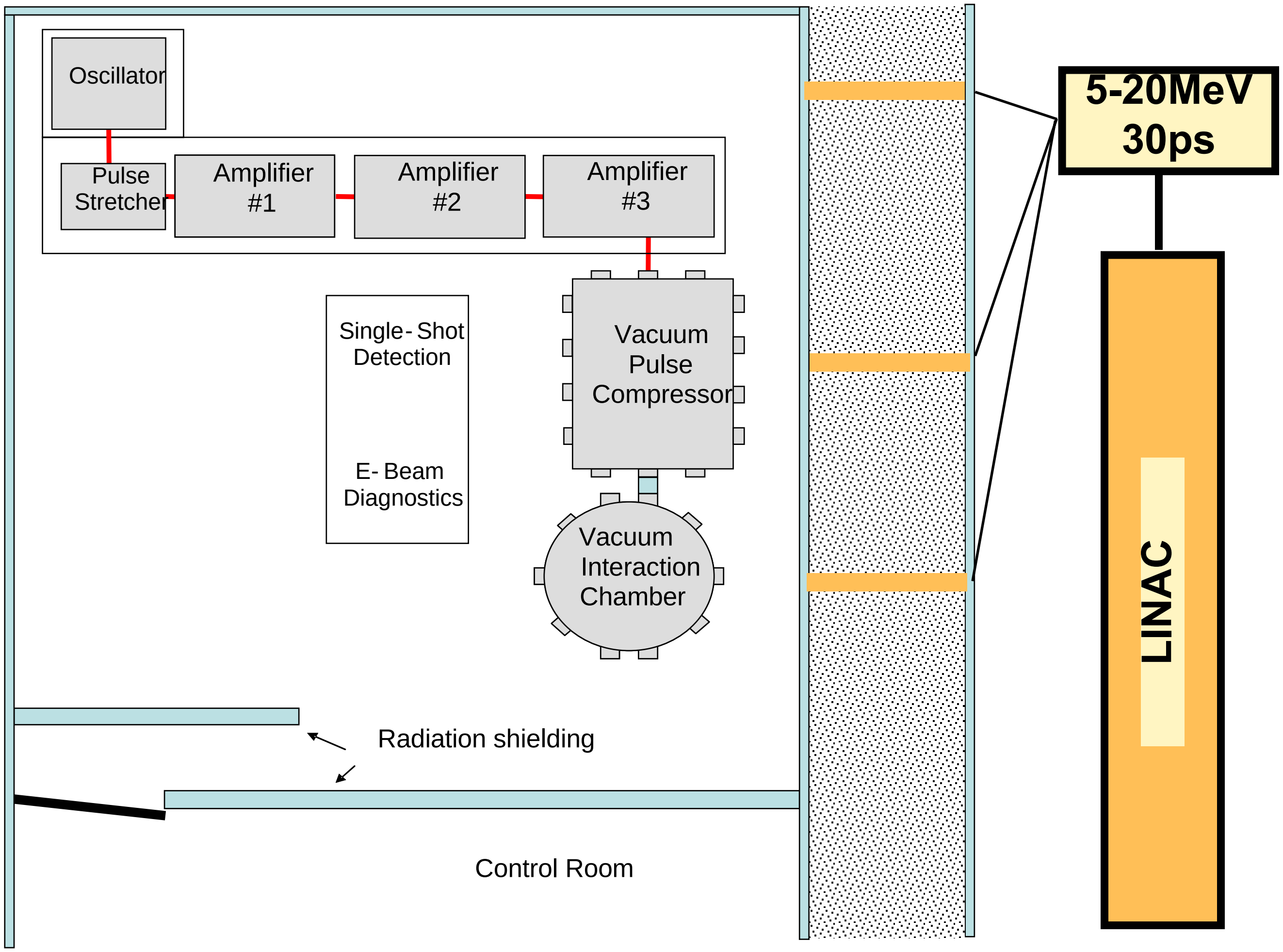
Requires

- $>10^{18}$W/cm^2

$\Rightarrow$ terawatt laser system
e.g., .5J in 50fs = 10TW

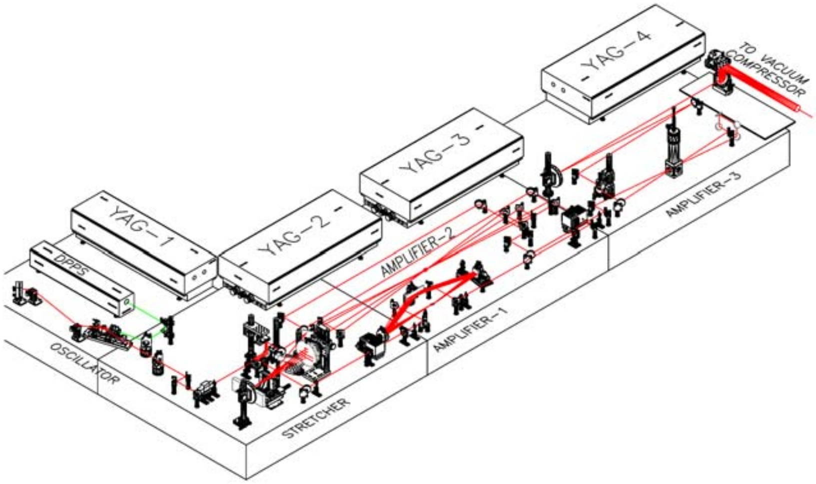
Pulse charges as high 1-5nC have been achieved using T^3

Terawatt Ultrafast High Field Facility



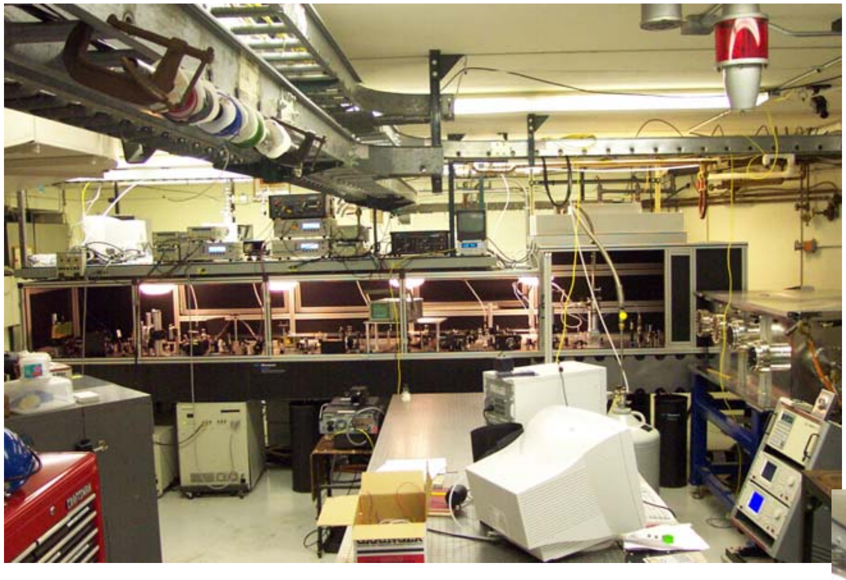
T^3 Specifications

	Wavelength	Rep.	Pulsewidth	Energy
Oscillator	780nm	100MHz	15fs	2nJ
Amp 1	800nm	10Hz	~350ps	2mJ
Amp 2	805nm	10Hz	~350ps	.35J
			30fs	**.15J (5TW)**
Amp 3	805nm	10Hz	~350ps	1.3J
			30fs	**.6J (20TW)**



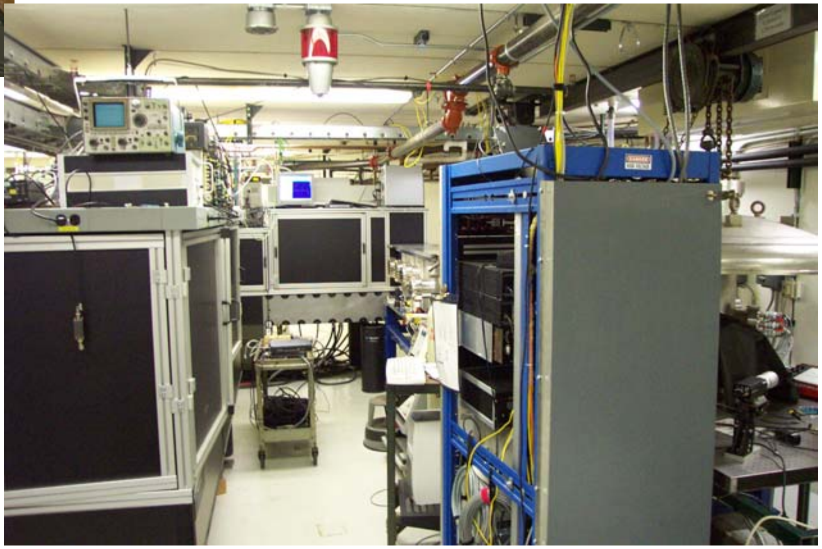
Future upgrade will increase the power to 50TW

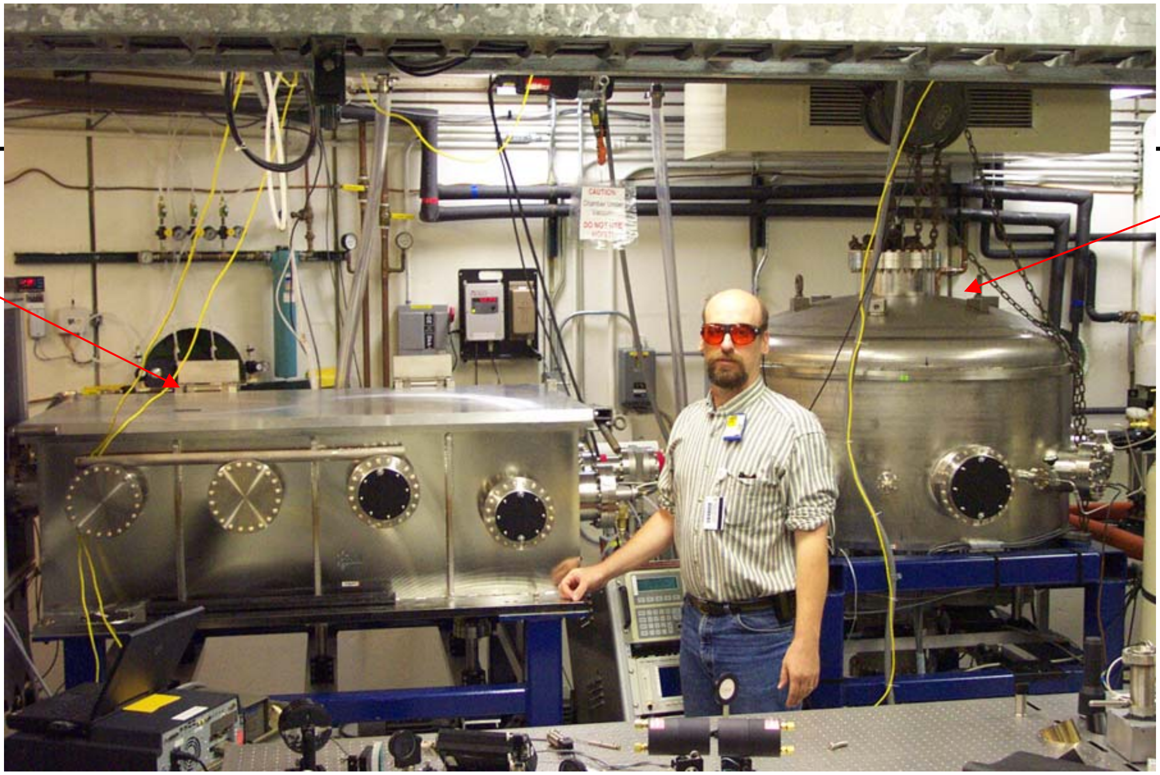
TUHFF



Sometime ago

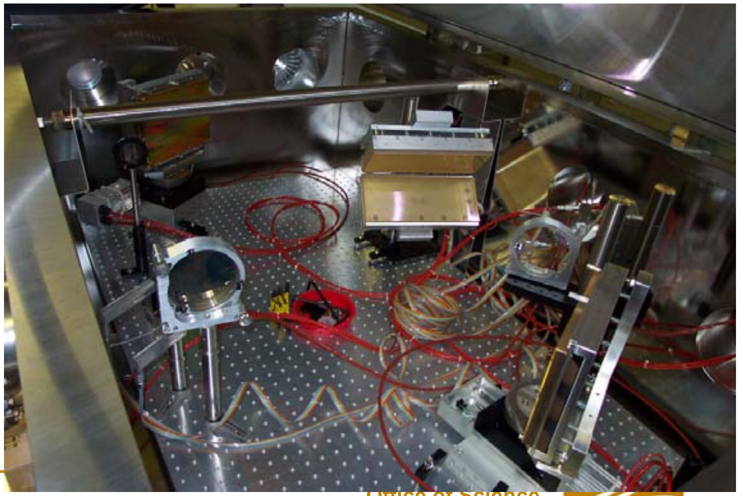
Present



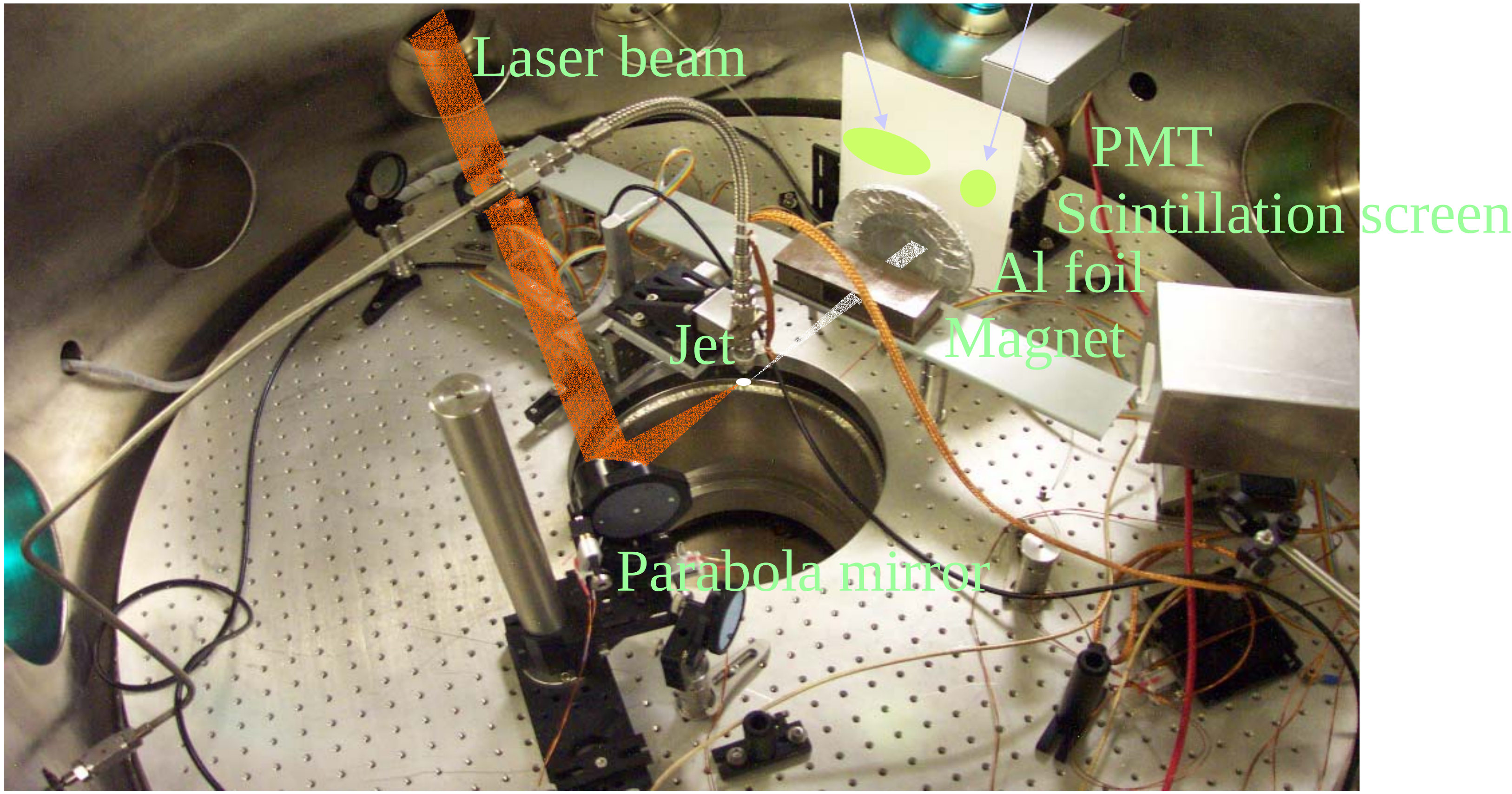


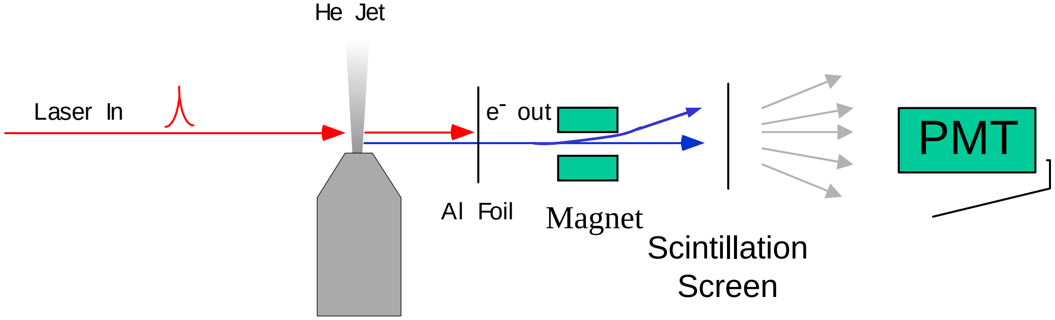
Vacuum Compressor

Target Chamber



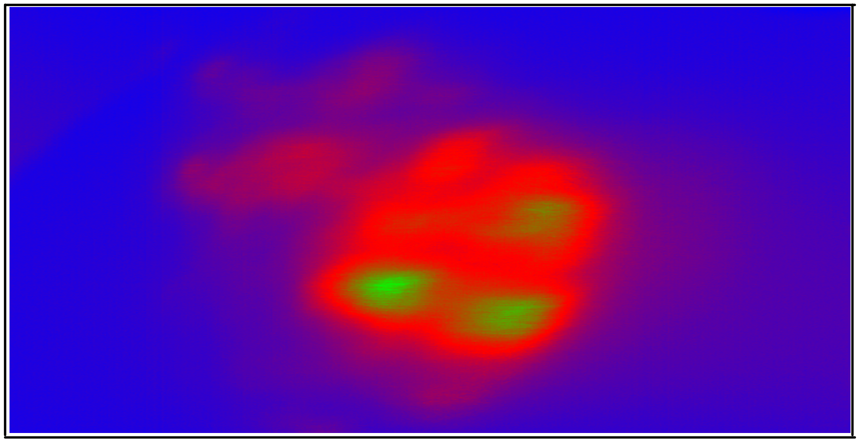
Laser Generation of Electron Pulses



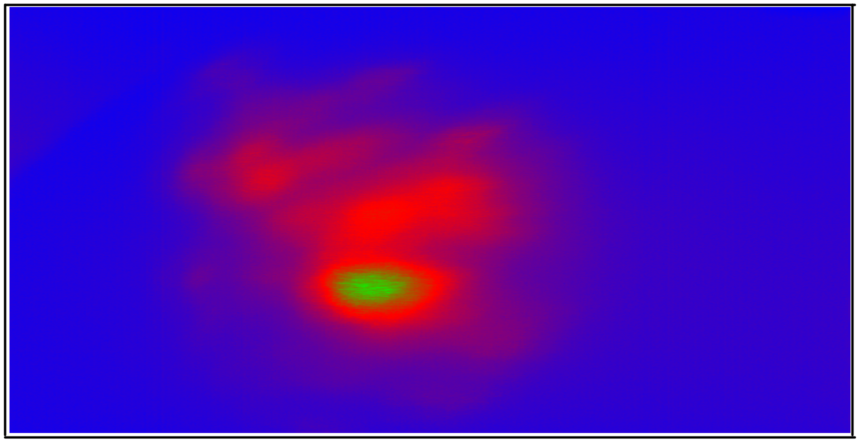


Electron Beam Spatial Profile

2TW

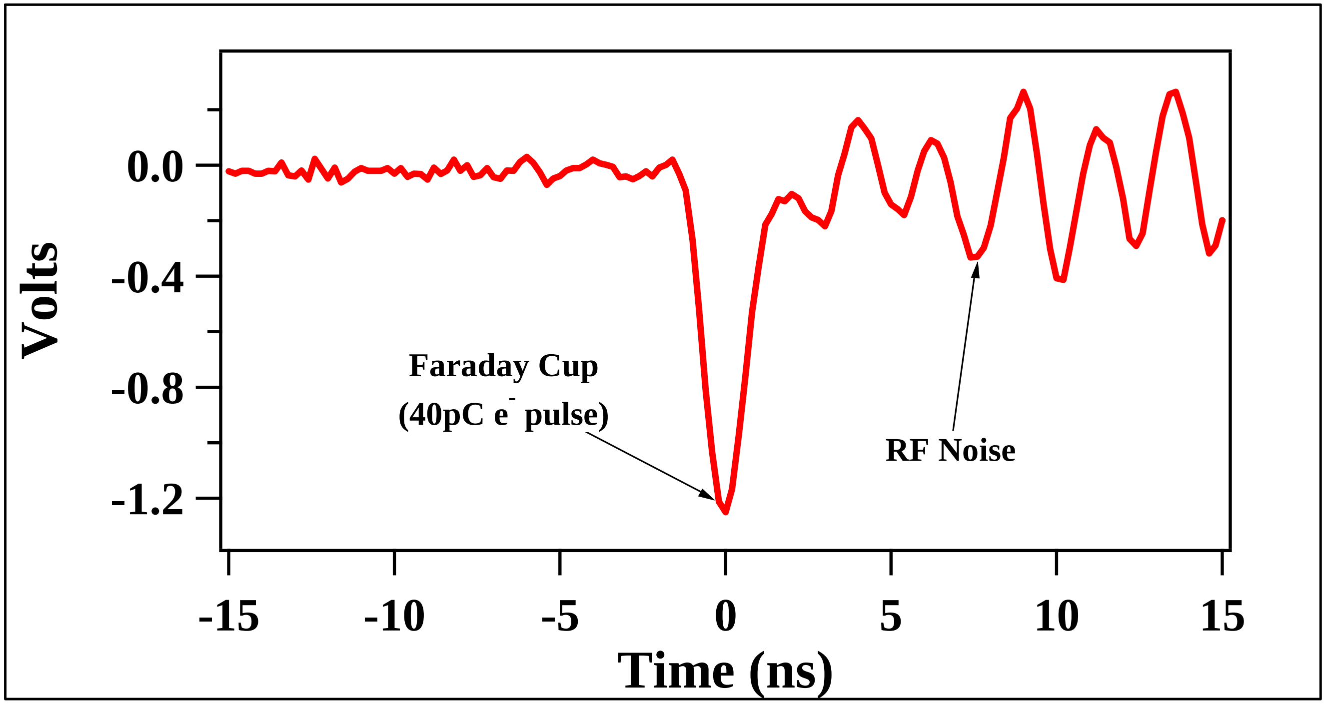


7TW



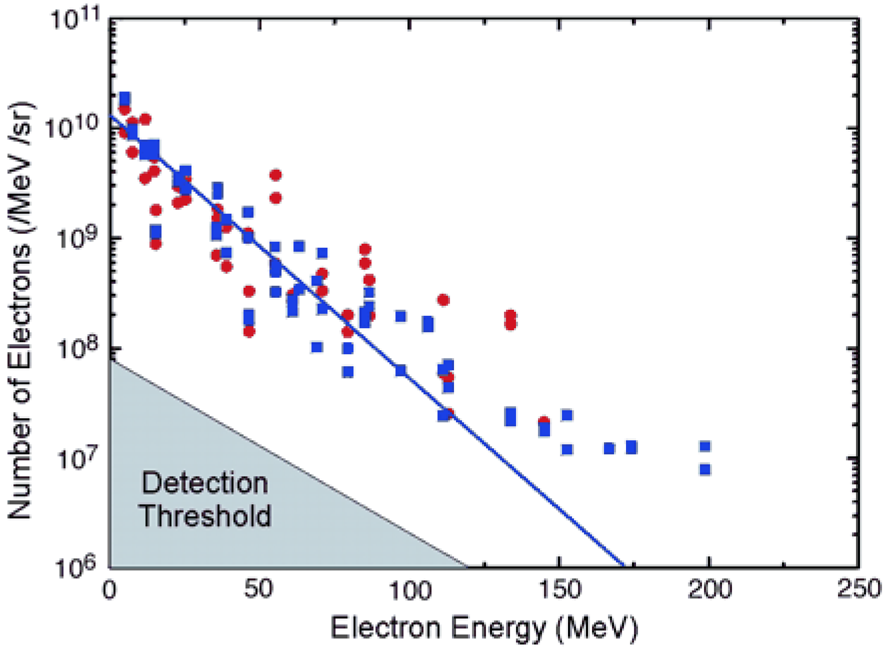
The full angle beam divergence goes from ~15° at low power (2TW) to ~3° at higher power (7TW). At the highest laser power (23TW) the divergence is expected to be on the order of 1°.

Measurement of Charge



The typical charges that we have measured are 400-600pC enough to start experiments with 2-5ps resolution!

Electron Energy Spectrum



Malka et. Al, Science 298 (2002) 1596

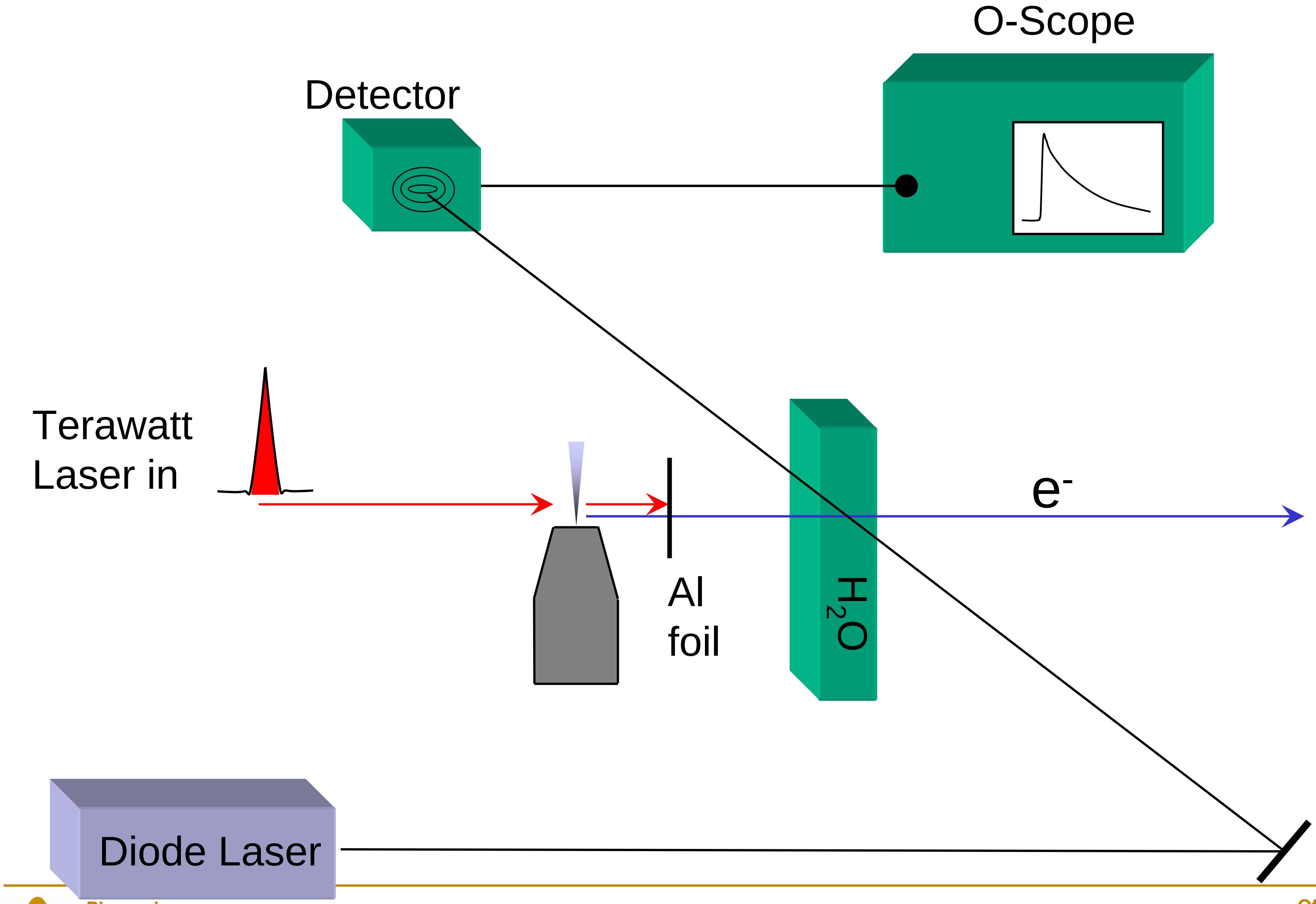
Large energy dispersion is a definite disadvantage
Dispersion = .5ps/cm

Monochromatic e$^-$ beam, low divergence - V. Malka

Pulse Radiolysis with T^3

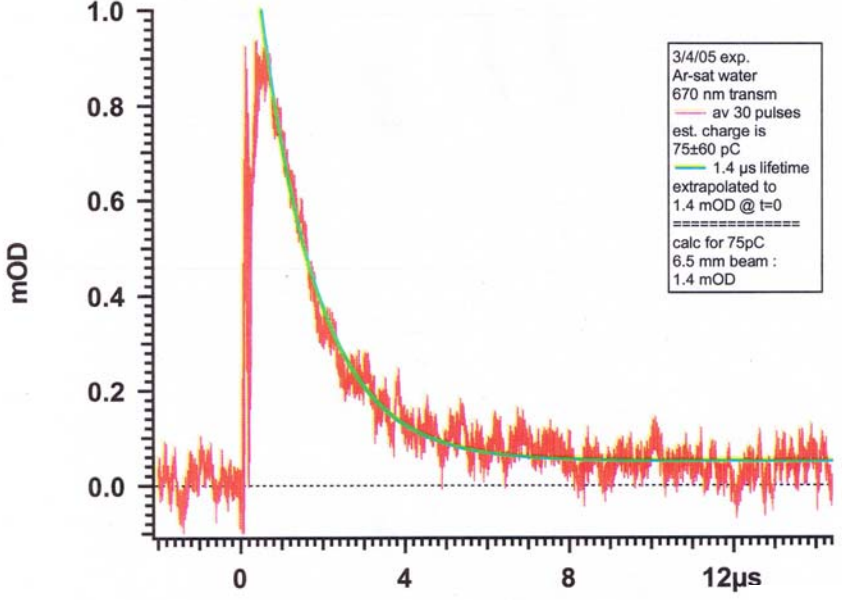
- Have enough charge to do electron pump optical probe measurements, but…….
- Current S/N is not good enough interpret quantitatively
- Long acquisition times are difficult because the sample is close to the jet
- Need to set-up an easier experiment to optimize picosecond measurements

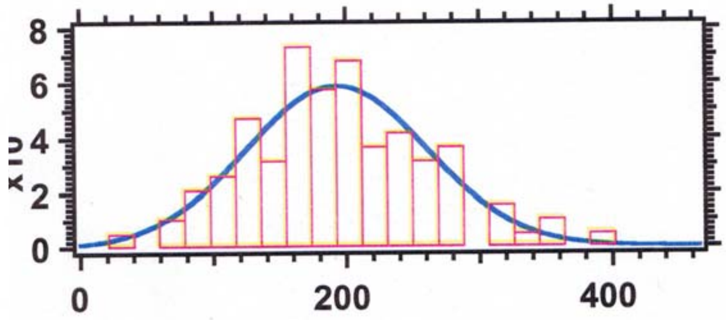
Optimization of detection



Statistics

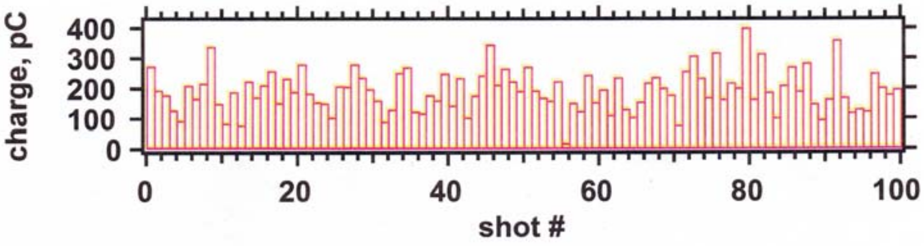
$e^-_{aq} + O_2 \rightarrow O_2^-$





Charge pC

Need to normalize pump-probe measurements to the pulse charge



Summary

- Primary processes in high energy chemistry are important have not been studied experimentally-also need theory
- Photoionization Experiments => primary events are fast, complex, do not reproduce spurs, but provide some insight
- TUHFF laser system (>20TW) has been constructed in the Chemistry Division and has successfully accelerated electrons to energies of several MeV
- Currently, pump/probe measurements on water

Acknowledgments

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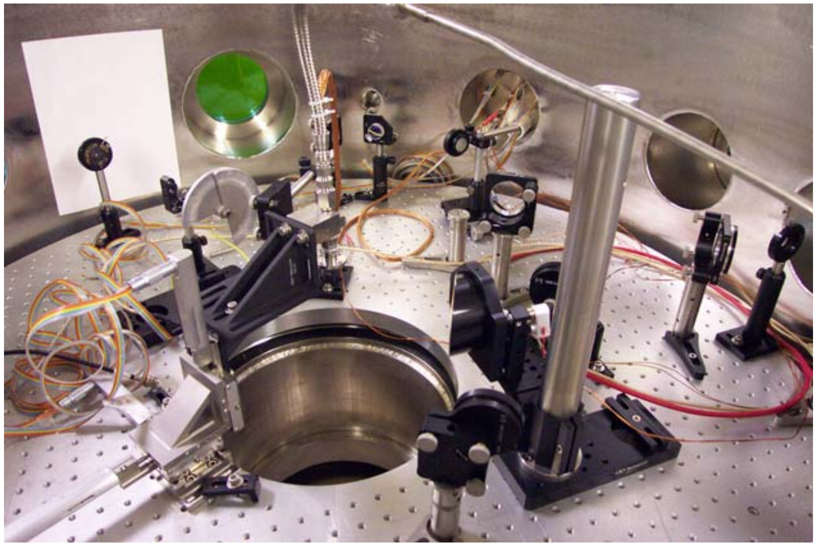

Inside of the TUHFF Target Chamber