Probing high local concentrations in track structure of high LET radiolysis

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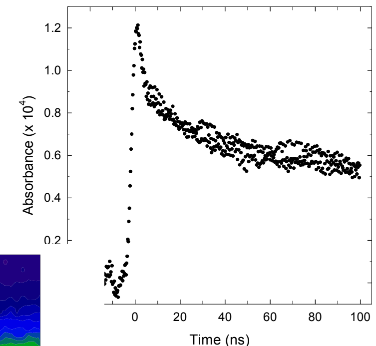
Introduction

Long term…

From stochastic system
… to deterministic solution.

Chemical kinetics



H_2

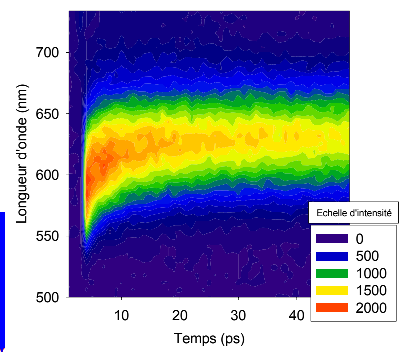
Corrosion

Solubilization (Waste management)

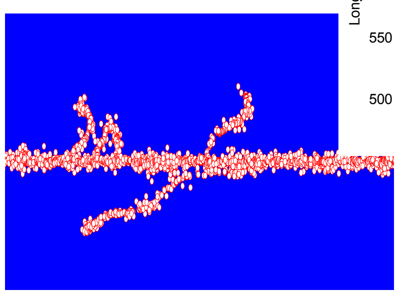
Radiobiological effects

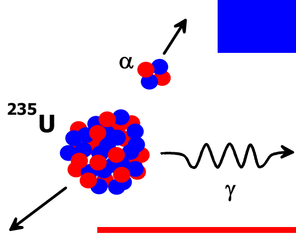
…

Solvation



Ionization tracks





Chemistry of solutions

Local effect

Heterogeneity

Diffusion of species

time

0 | 10^{-15} | 10^{-12} | 10^{-9} | 10^{-6} | 1s à 10000 years

LET

$$LET = -\left(\frac{\mathrm{d}E}{\mathrm{d}x}\right)_{\mathrm{elec}}$$

unit : keV/μm ou eV/nm

Bethe formula : $$-\frac{\mathrm{d}E}{\mathrm{d}x} = \frac{2\pi e^4 Z^2 Nz}{E}\frac{M}{m}\ln\frac{4E}{I}\frac{m}{M} = \frac{Z^2}{\beta^2}\frac{4\pi e^4 c^2 Nz}{m}\ln\frac{2m\beta^2}{Ic^2}$$

Orders of magnitudes in water:

e- (MeV) ou γ	: 0.2 – 0.5 eV/nm
β- (18 keV du ^{3}H)	: 2.7 eV/nm (mean value)
C^{6+} (1 GeV)	: 30 eV/nm
Ar^{18+} (3 GeV)	: 250 eV/nm
He^{2+} (5 MeV)	: 130 eV/nm (mean sur 35μm)

Dose : Locally deposited energy for 1 kg of water
unit : Gy ou J/kg

Radiolytic yield (g) : conc. of produced (or consumed) species / dose
unit : mole/J

LET effect on the radiolytic yields

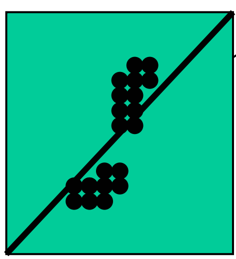
$$H_2O \xrightarrow{\text{ionizing ray}} OH^\bullet, H^\bullet, e^-_{aq}, H^+, HO_2^\bullet, H_2O_2, H_2$$

γ ^{60}Co rays LET = 0.25 eV/nm			accelerated ions LET > 0.25 eV/nm	
e^-_{aq}	2.7×10^{-7}mol/J	↘	e^-_{aq}	< 2.7×10^{-7}mol/J
OH•	2.7×10^{-7}mol/J		OH•	< 2.7×10^{-7}mol/J
H•	0.6×10^{-7}mol/J		H•	< 0.6×10^{-7}mol/J
$HO_2^\bullet$	0 mol/J		$HO_2^\bullet$	> 0 mol/J
H_2	0.45×10^{-7}mol/J	↗	H_2	> 0.45×10^{-7}mol/J
H_2O_2	0.7×10^{-7}mol/J		H_2O_2	> 0.7×10^{-7}mol/J

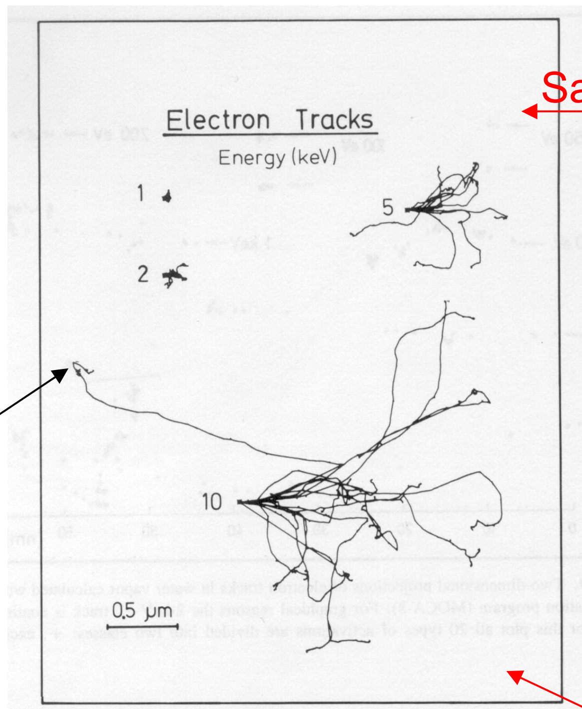
Structure of the tracks and LET values

When LET do not explain the results… how to access to structure of tracks?

Atomic number (Z) — Energy (β)

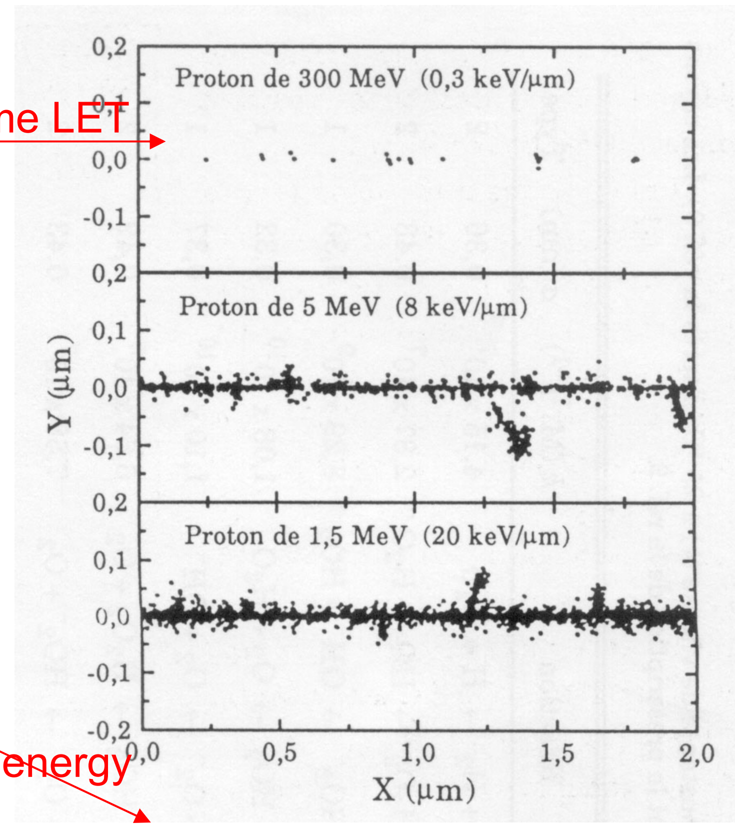


Spurs



Same LET

Same energy



Freeman, G.,1987,
« Kinetics of non homogeneous processes »

Cobut *et al.*, J. Chim. Phys., 1995, (93) 93-100.

What is the chemistry with high concentration?
How to probe local high concentration at fs-ps?

Impossible to use « final products analysis » by using scavenging method

The concentration of scavengers should be > 5M

Possible direct effect on the solute

So, it remains real time methods : transient species

- Pulse radiolysis (absorption, fluorescence…), « μs-ns-ps limited »

Pump probe spectroscopy

Hydrated electron
An ideal probe of the track structure

Formation in the fs-ps time range

Properties: λ_{max} = 720 nm, ε = 20000 M^{-1} cm^{-1}

Many studies existe

Few time-resolved studies at high LET, direct detection

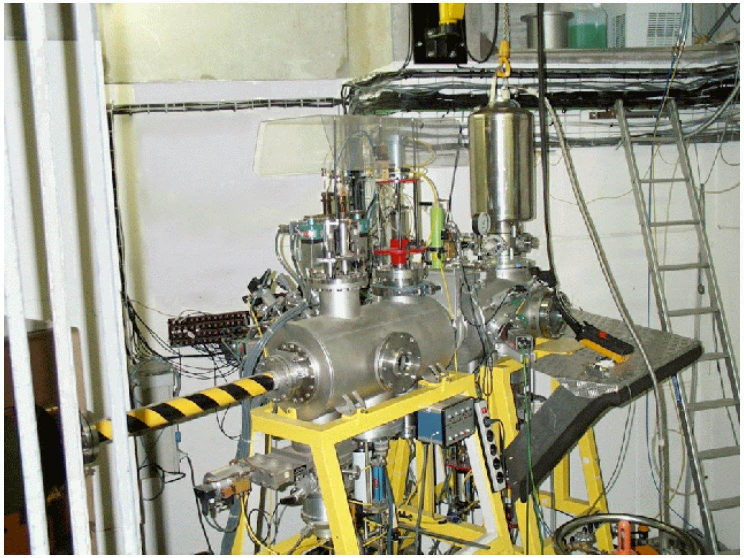
helions 10μs: Sauer et *al.,* 1977;

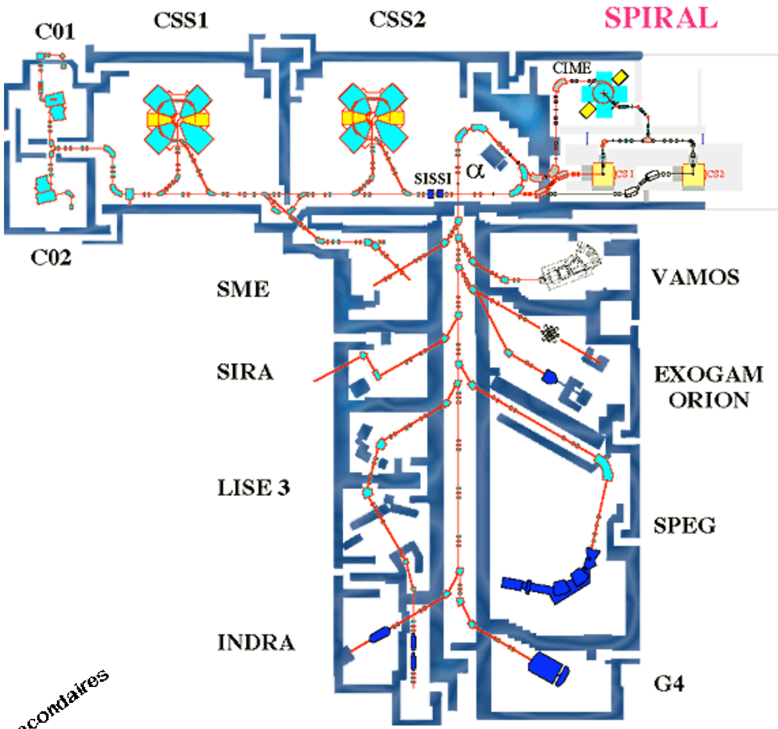
protons 1ns: Buxton et *al.*, 1981

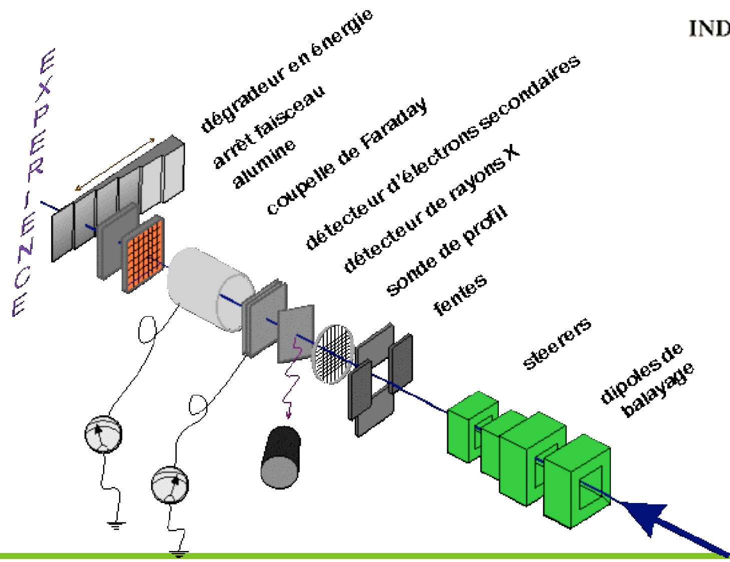
But: low concentrations at high LET

But: high concentration at early times

High energy ions at GANIL

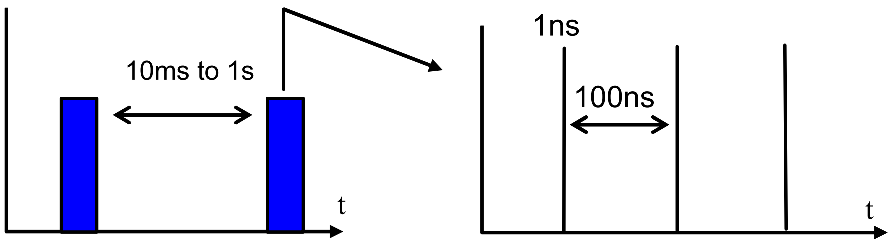


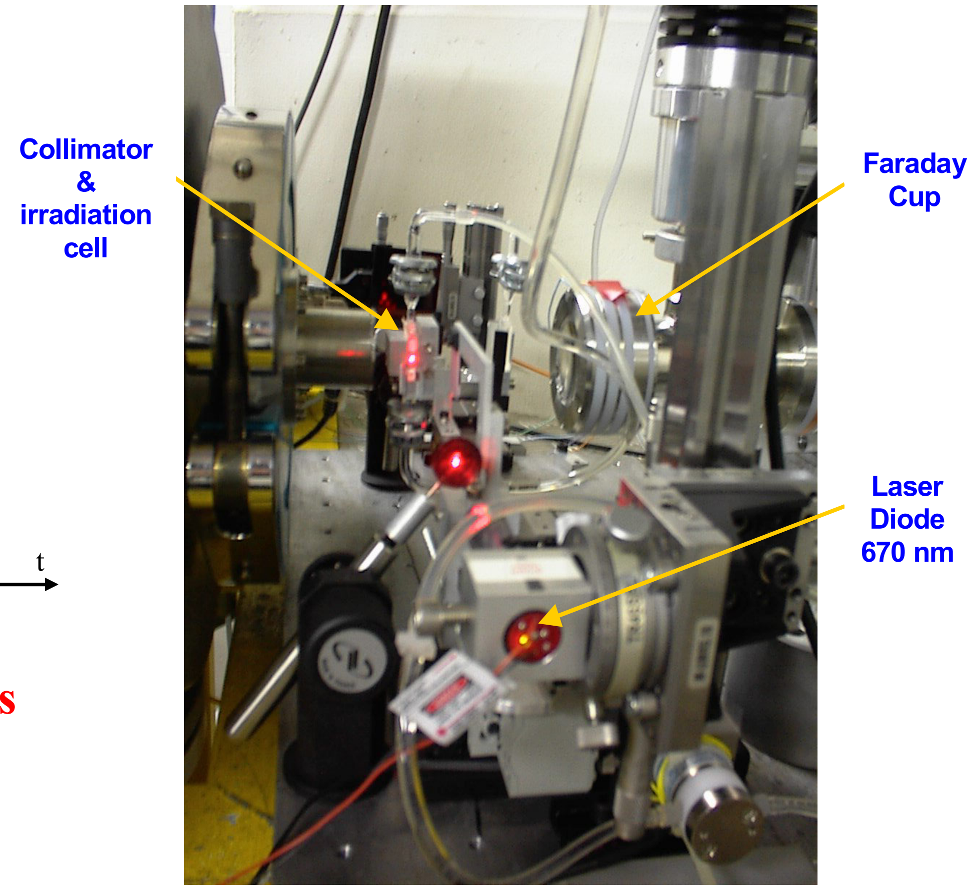




Pulse radiolysis setup

Temporal structure of the beam





Duration of the pulses : 1ns or > 5µs

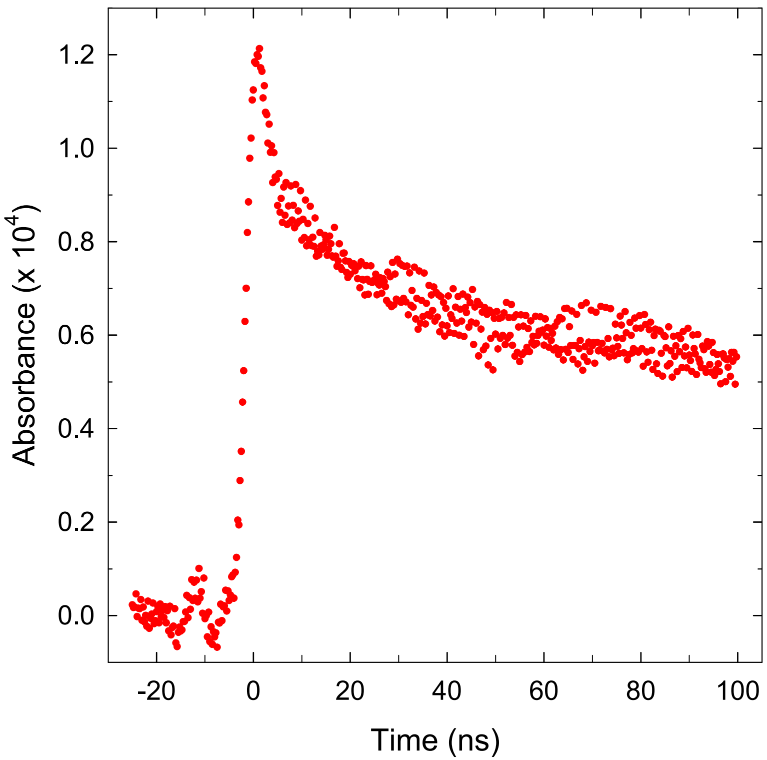
Formation and decrease of hydrated electron

Experimental conditions

- C^{6+}, 95 MeV/amu, 27 eV/nm
- 1 ns, 1 kHz, 10^6 acquisitions
- De-aerated pure water
- dose = 6 10^{-3} Gy/pulse

Results

- Low levels of absorption / conc.
- Fast reactions in 10^{-7}s
- $g_{1ns}(e_{aq}^-)$ = 4.5 10^{-7} mol/J



Baldacchino et al., NIMB (2003) *209*, 219-223

With 2 ions with same velocity

Incident ions

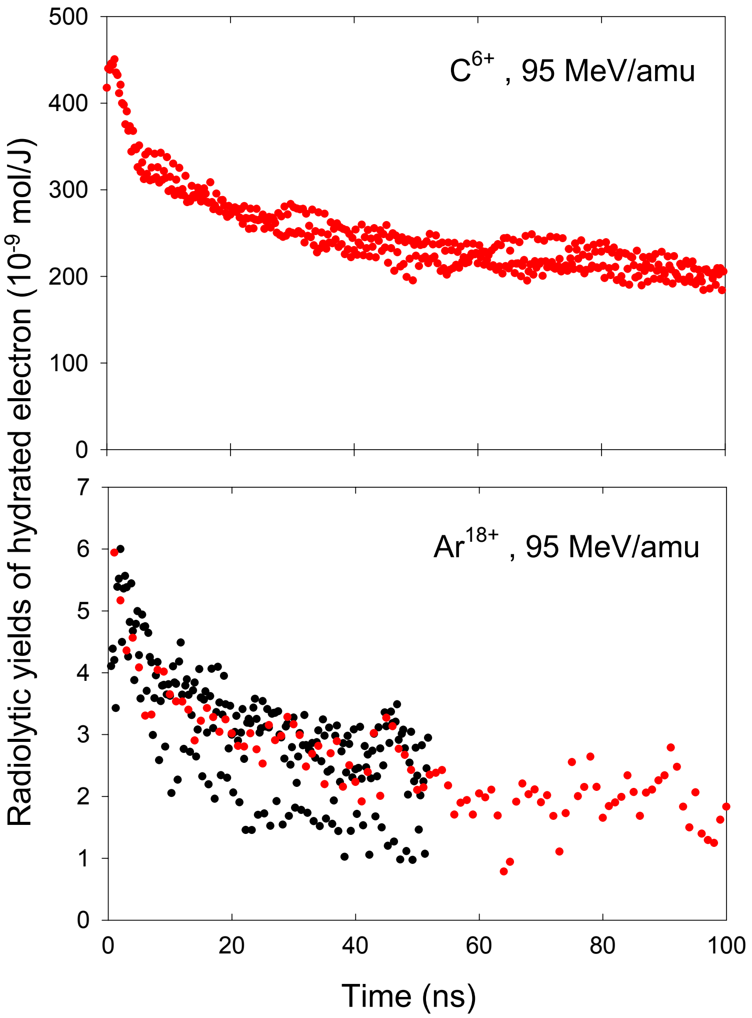
- C^{6+},95MeV/amu, 27eV/nm
- Ar^{18+},95MeV/amu, 250eV/nm
- => same velocity: $\beta = v/c = 0.32$
- => TEL[Ar^{18+}] # 10 × TEL[C^{6+}]

(i.e.: Bethe formula)

- => Z^2[Ar^{18+}] # 10 × Z^2[C^{6+}]

Results for e^-_{aq}

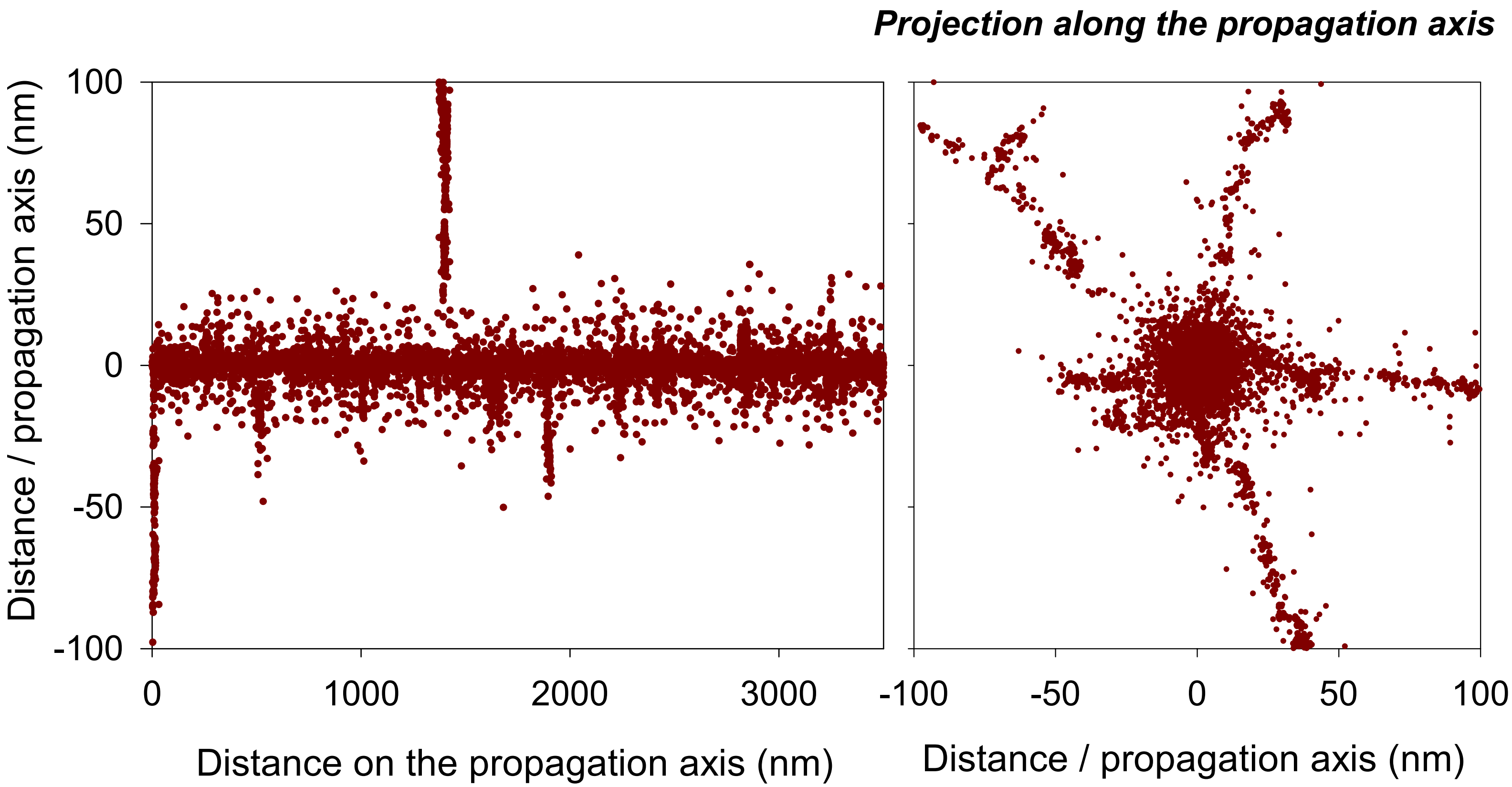
- g_{1ns}[C^{6+}] # 100 × g_{1ns}[Ar^{18+}]
- In 100ns,
  - g/3 for Ar^{18+}
  - g/2 for C^{6+}



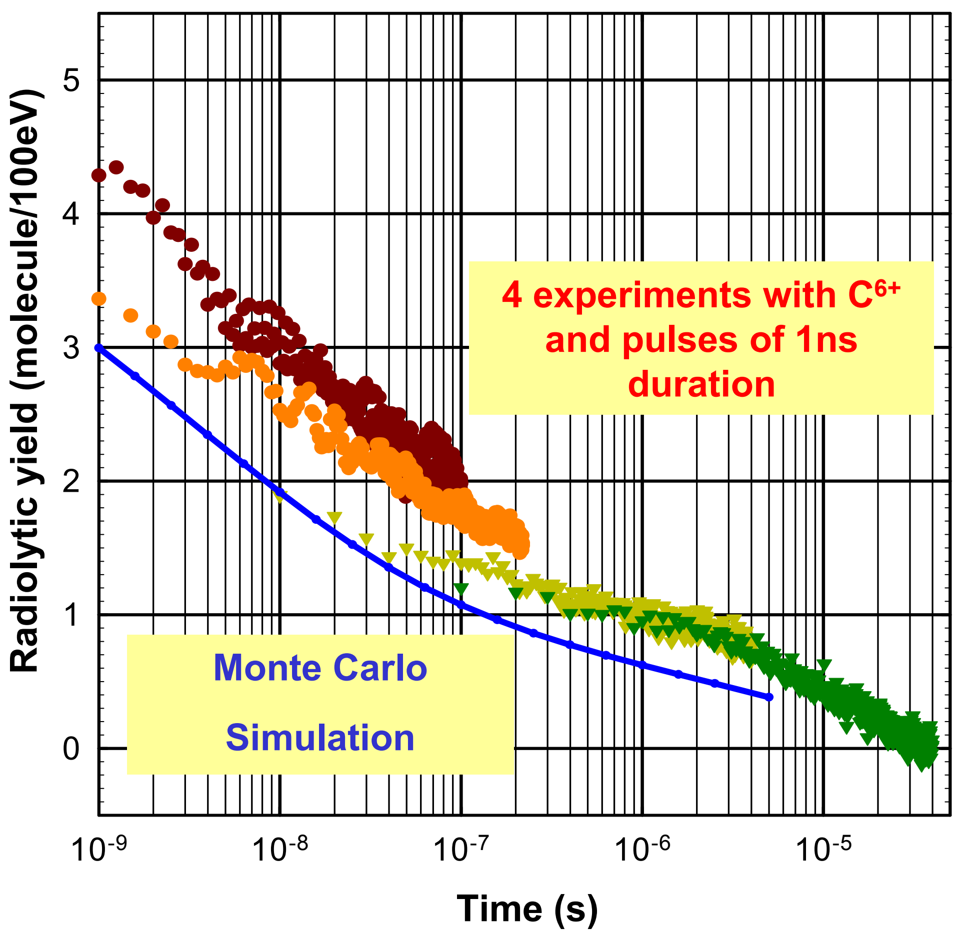
Baldacchino et al., Chem. Phys. Lett. (2004) *385*, 66-71

Monte Carlo simulation

C^{6+} ions, position at t=1ps



Comparaison Experiments / MC Simulation



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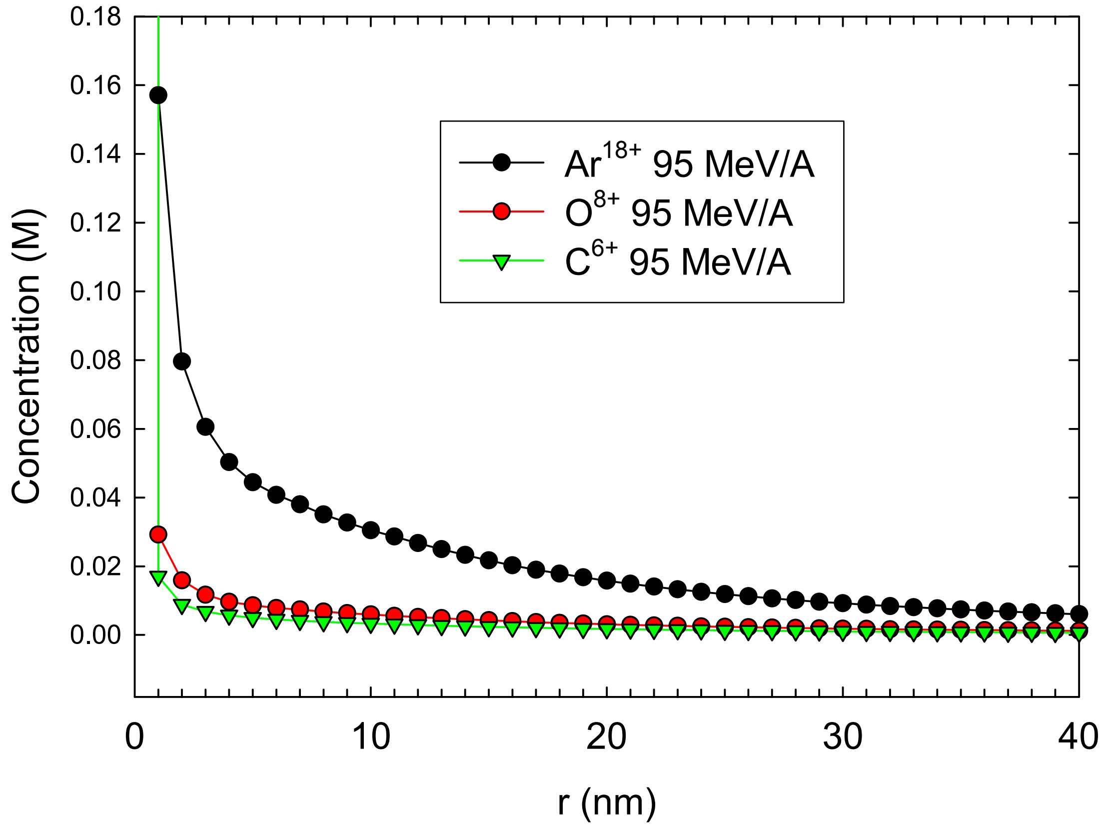
Concentrations of free radicals in the tracks



About hydrated electron

- **Results at 1ns:**

  With C^{6+} to Ar^{18+}

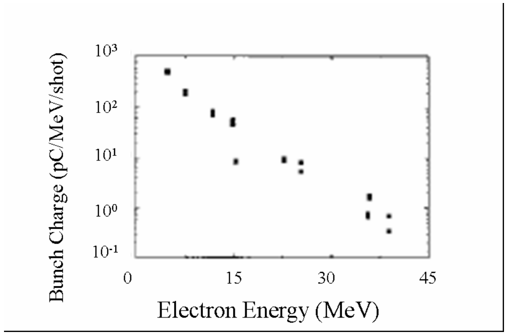
- **1ns – 100ns : complex kinetics**

  Heterogeneous processes

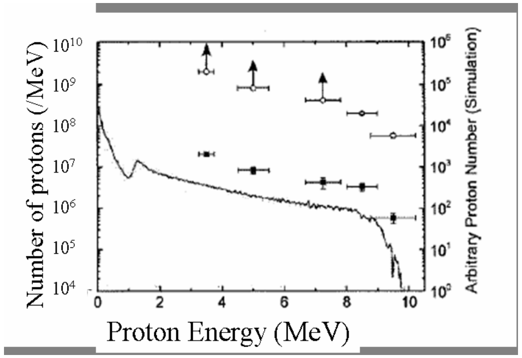
  High concentrations at early times

- **Necessity to probe it with fs-ps pulses**

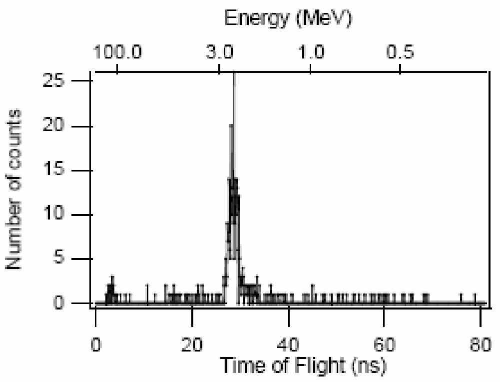
Electrons



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

Projects of pulsed high energy ions accelerators

Femto-pico proton / neutron

- **Based on T^3-laser at DRECAM/Saclay (Cf Stanislas Pommeret)**
- **Pump probe experiments (« single shot » method?)**

Over pulsed accelerators
Long term projects and intermediate time-resolution

Cyclotrons (He^{2+}) : ns – μs (GANIL, Subatech Nantes)

Developments of Linear Accelerator of p^+, d^+ :
high energy and 100 ps (GANIL)