Swift heavy ions, ices and astrophysics

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e de Recherche sur les Ions, les Matériaux et la Photonique

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Ices?

Ions? In space, in the lab (GANIL) and in matter.

Laboratory simulations :
Several examples

Water : compaction and amorphisation

Role of CR : CO ice

Gaz mixture and complex molecules

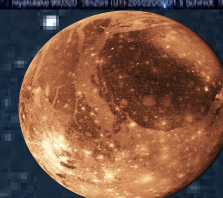
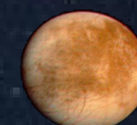
Perspectives : IGLIAS, next?

Astrophysical Ices …

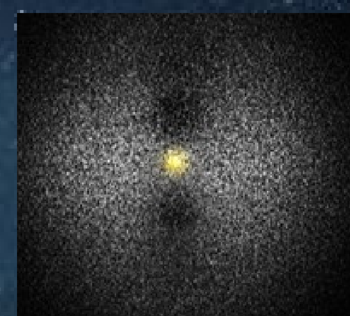
- Comets



- Giant Planet's Moons
  (Europa, Ganymede, …)

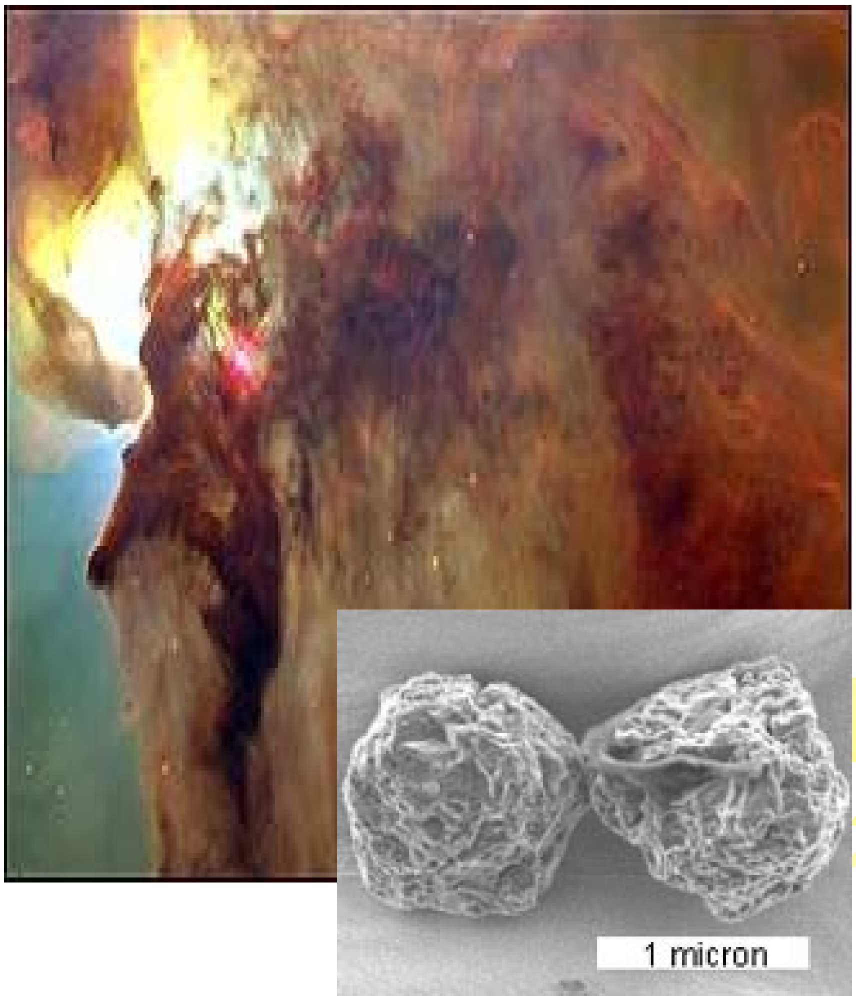
- Dust Grains

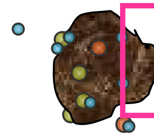




Rings Dense Interstellar Clouds

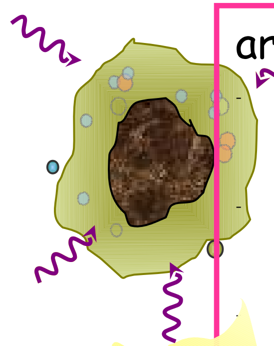
(birthplaces of suns and planets)

Interstellar dust grains (dense molecular clouds)





... covered with thin layers of ices (H_2O, CO, NH_3, ...)



are exposed to
- cosmic rays;
  (protons, helium, heavy ions)
- stellar wind
  (H, He, C, O, S ...)
- UV photons
- electrons

irradiation leads to ...

Radiolysis

fragmentation/destruction

formation of molecules
(radiation chemistry)

Desorption / Sputtering

Compactation / Amorphization

- Primary Cosmic Rays are very energetic (103 to 1022 eV) charged particles that traverse outer space

1 kJ

- Basically, they are:

- light ions: protons + deuterons (87%) and α particles (11%)
- heavy 4n ions : 12C, 16O, 20Ne, 24Mg, 28Si, 32S, 40Ar, 40Ca and 56Fe /(Ni)
- electrons (~1%)

[unstable ions or neutrals are excluded: neutrons, neutrinos, X-rays, γ rays]

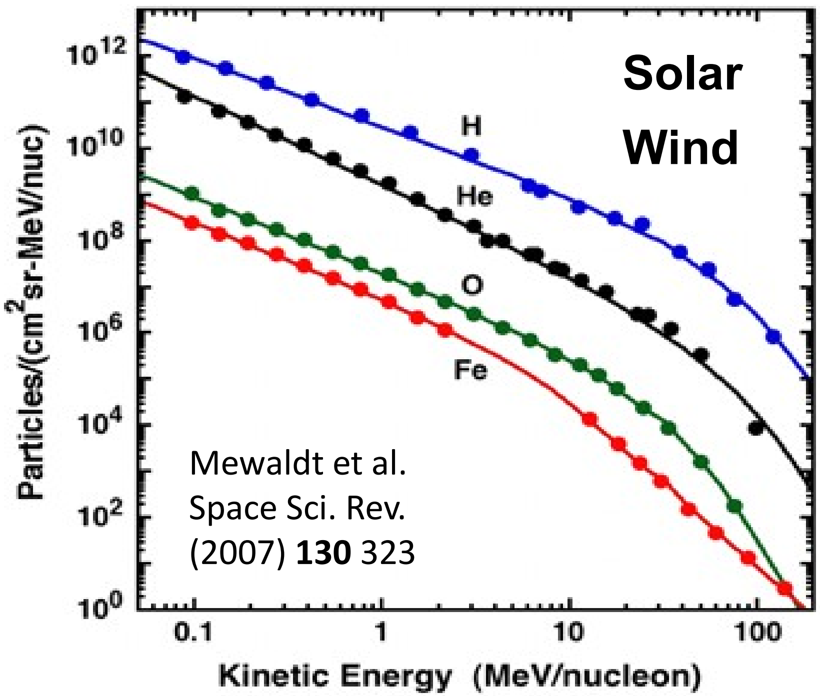
After collision with interstellar matter and atmosphere,

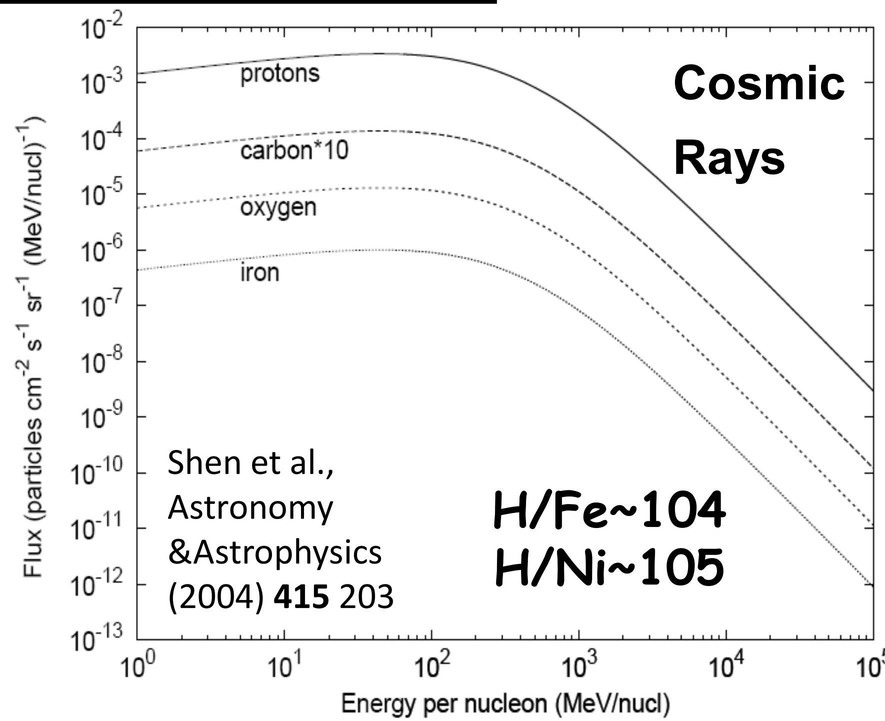
Secondary Cosmic Rays are formed. They are constituted by:

- Li, Be, B, neutrons (formed by spallation)
- pions, kaons, mesons, positrons and γ rays

Thanks to Enio

Concerning heavy ions in space:



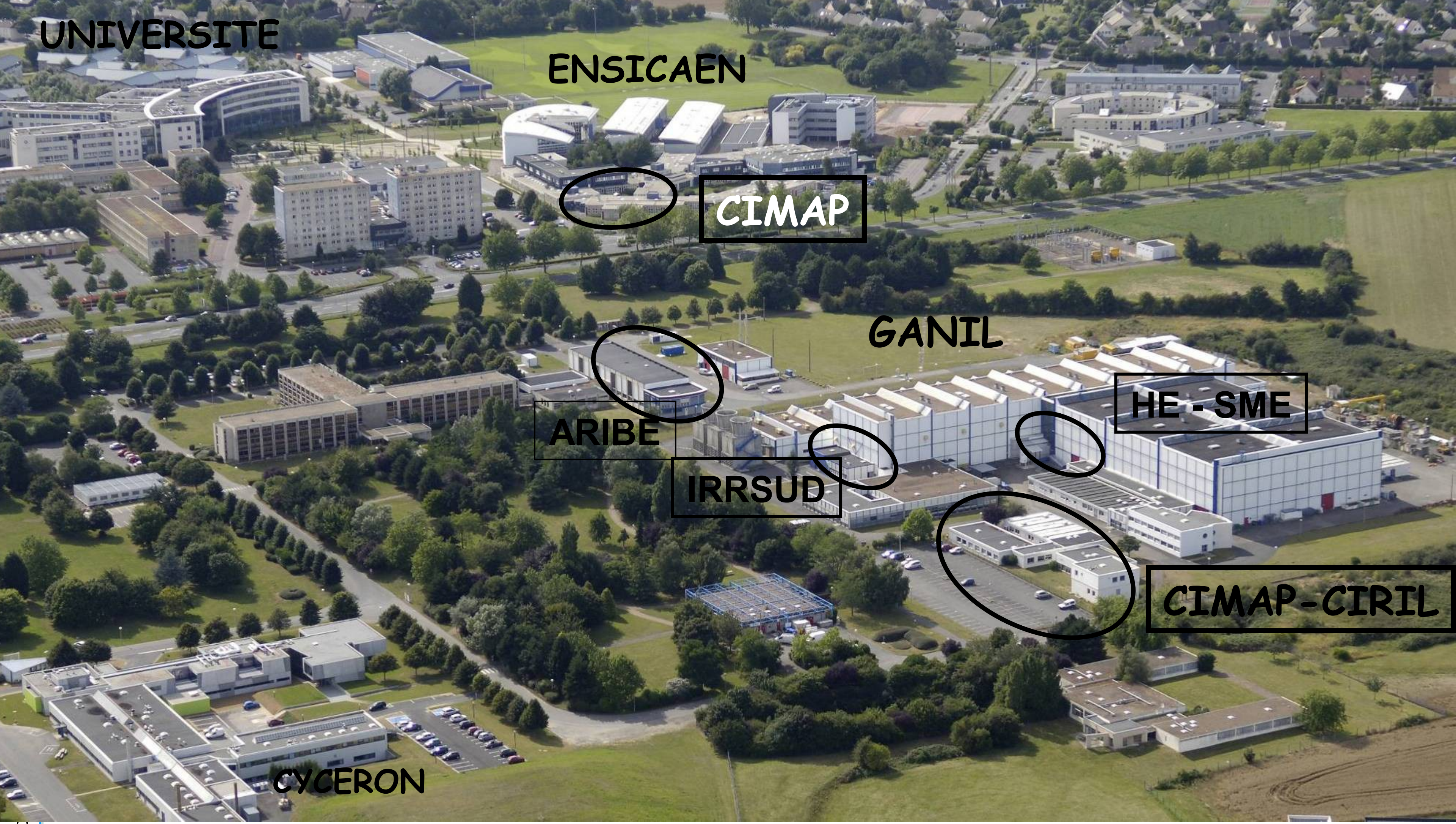


Heavy multiply charged Ions: - Large electronic energy loss **Se**

- Scaling laws: ***Sen*** with n ≈ ½,1,3/2,**2**, ... 4)
- Unexplained findings (gas phase CO in dense clouds...), few data
- Astrochemistry: origin of CO2 and H2SO4 on Europa, implantation.

- Shorter time for experiments…

Caen: a big accelerator of particles, GANIL



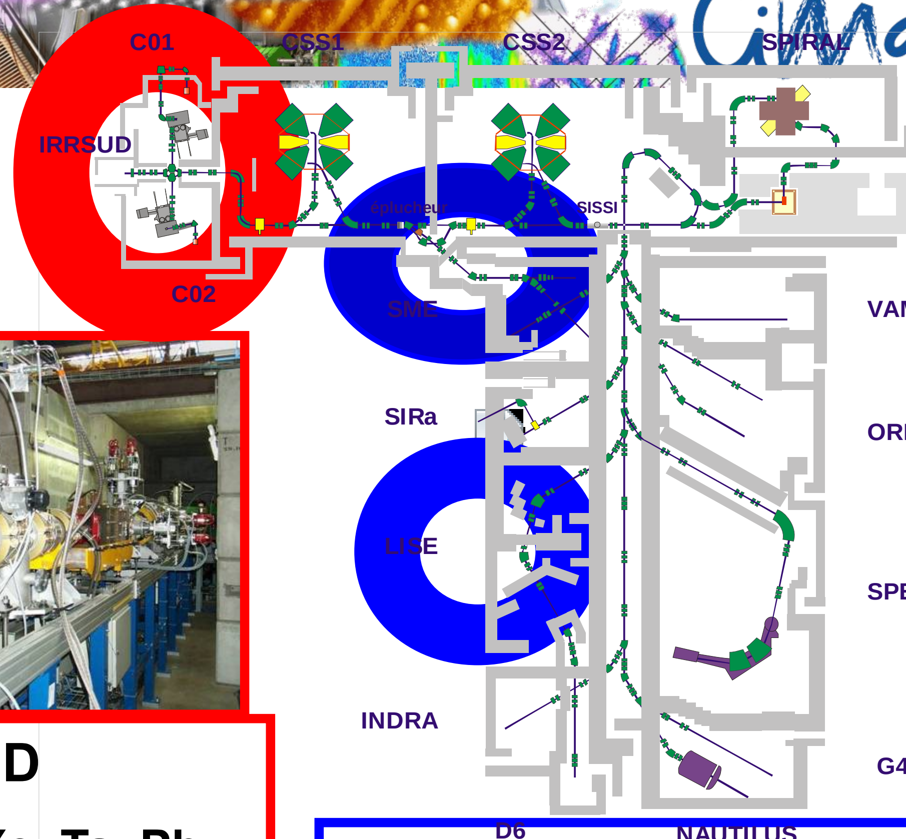
Du carbone à l’uranium, de l’eV au GeV
From Carbon to Uranium, from eV to GeV

User's Facility : you can apply for beam time!

Centre de Recherc

nom

HE, SME, IRRSUD



+ARIBE low energy multiply charged ions

He, C, O, S, Ar, Xe:

q keV

IRRSUD

O, Ni, Xe, Ta, Pb:

0.5 to 1 A MeV

High Energy: LISE

Fe, Ni: 70 A MeV

Medium Energy: SME

O, Fe, Ni, Kr: 5-13 A MeV

3 GeV for iron ion!

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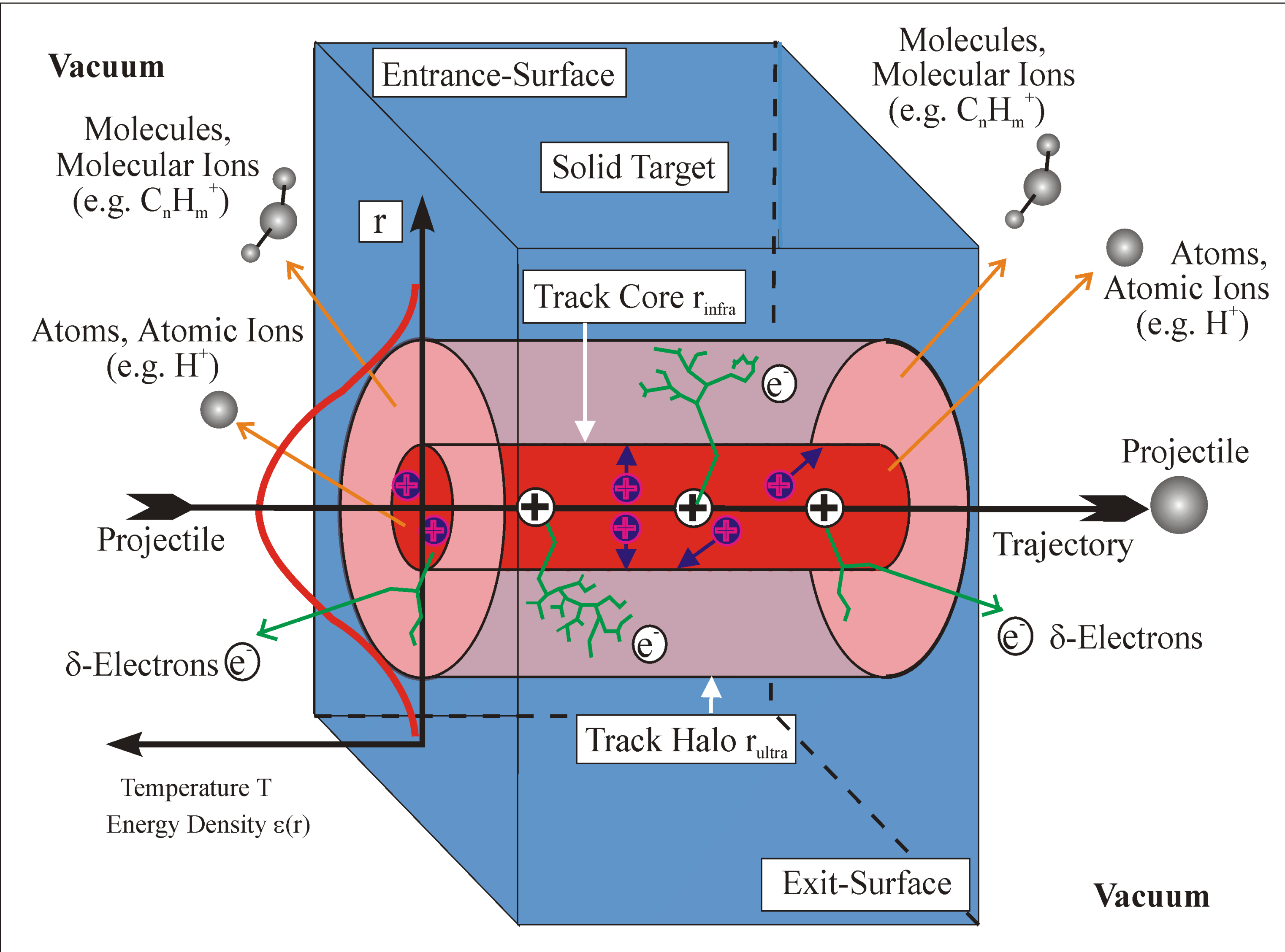
ions in condensed matter: nuclear tracks



Fast process
10-15s

1-30 MeV/µm
(1-30 KeV/nm)
Ve: 0 to 2Vp

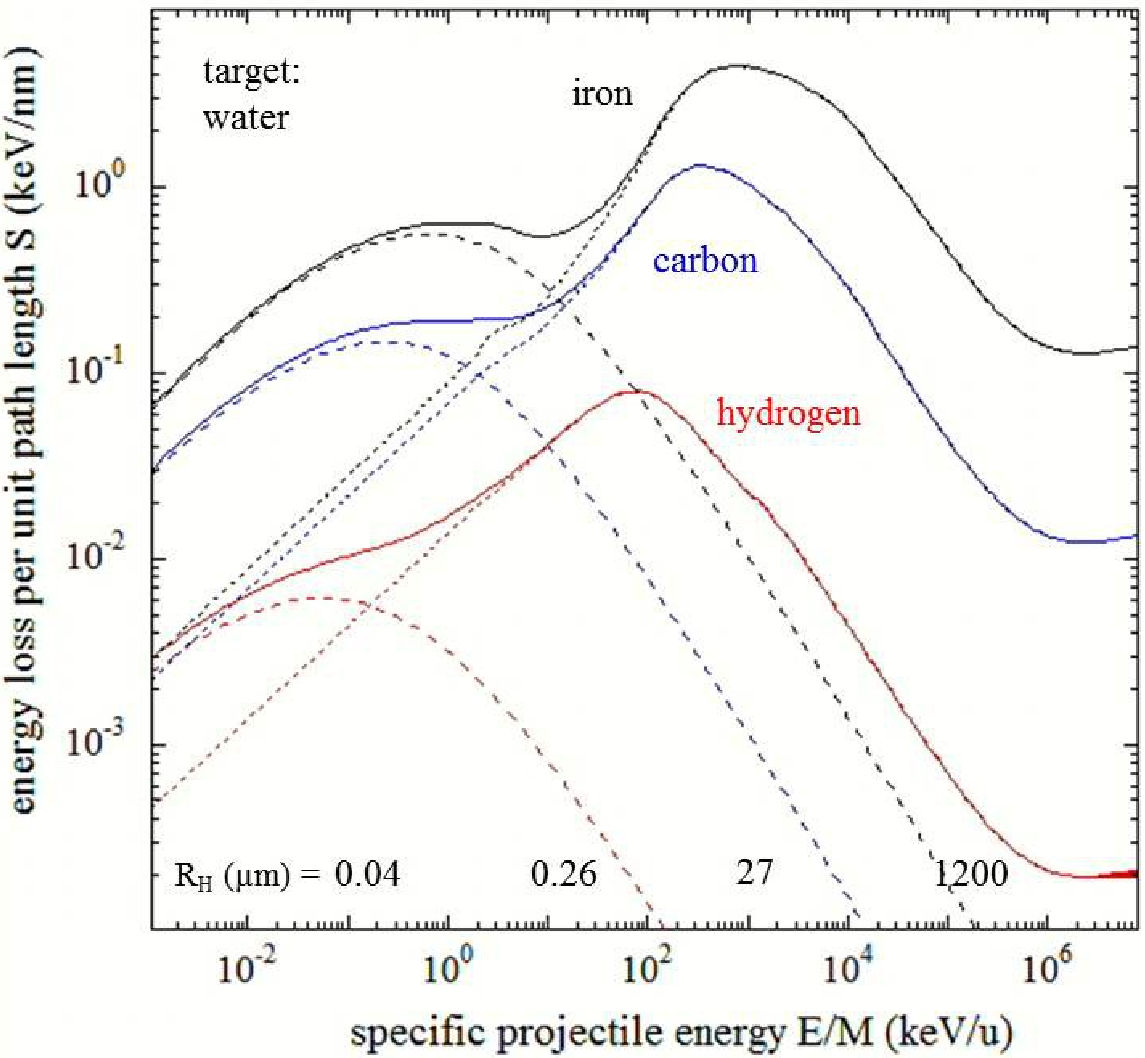
- Radicals

20000 K for
Picoseconde

Ysput :
104-105

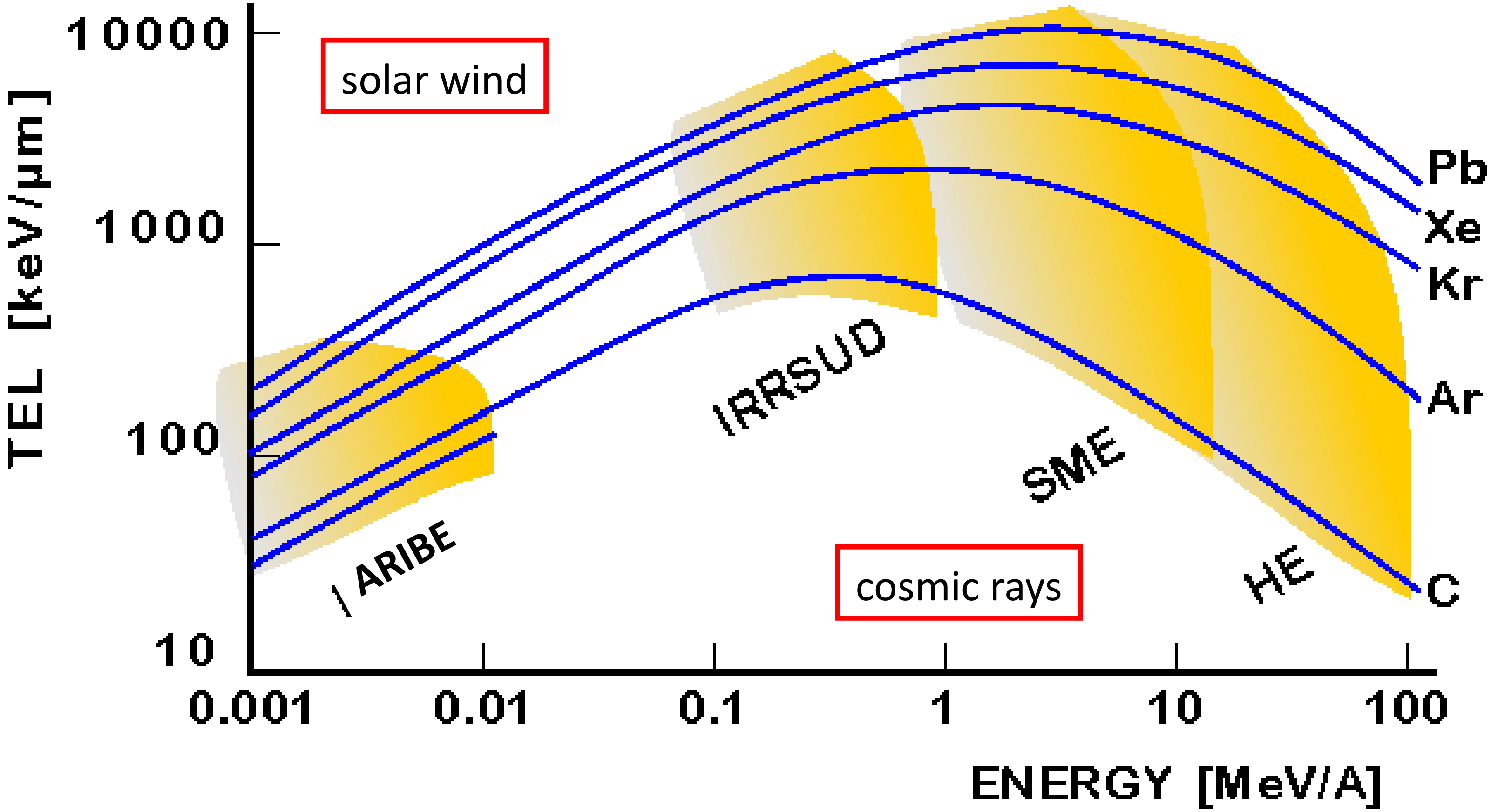
nom

For the incoming projectile:
The stopping power dE/dx :
Energy loss per lenght unit



Projectile	Se (keV/nm)
$^{58}Ni^{13+}$	3.0
$^{58}Ni^{11+}$	2.9
$^{64}Ni^{24+}$	2.0
$^{20}Ne^{6+}$	0.92
$^{16}O^{2+}$	0.79
$^{16}O^{5+}$	0.67

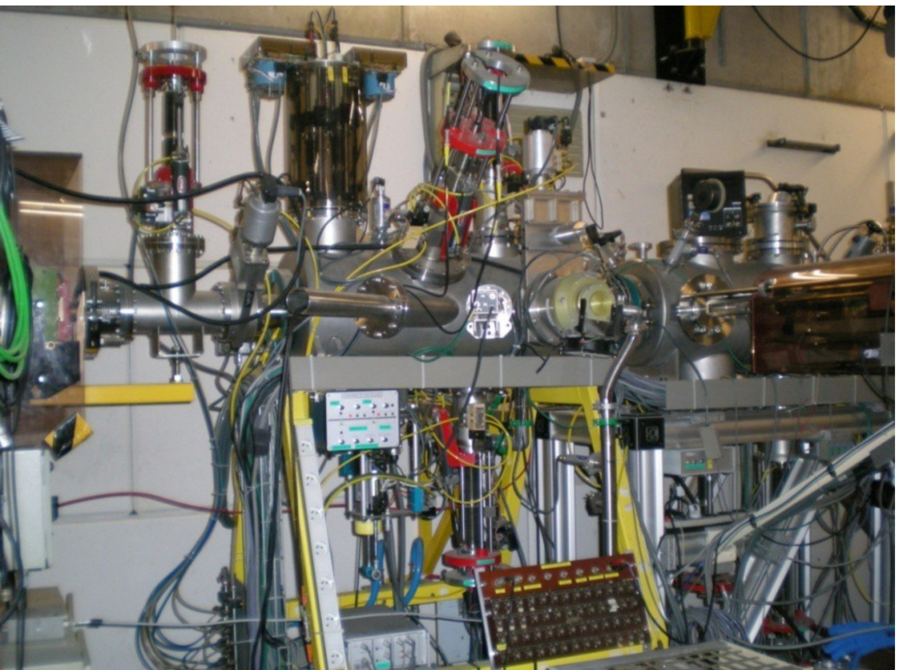
H+(100keV) Se=0,08 KeV/nm

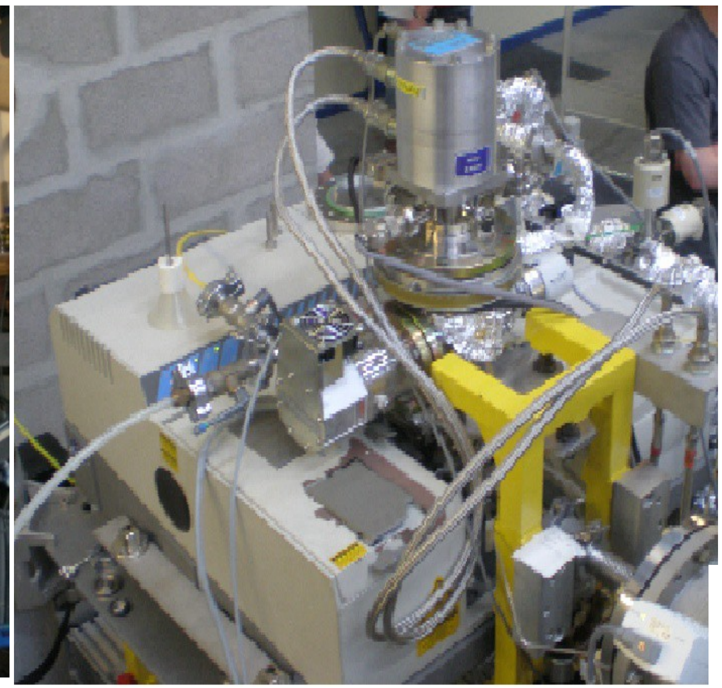


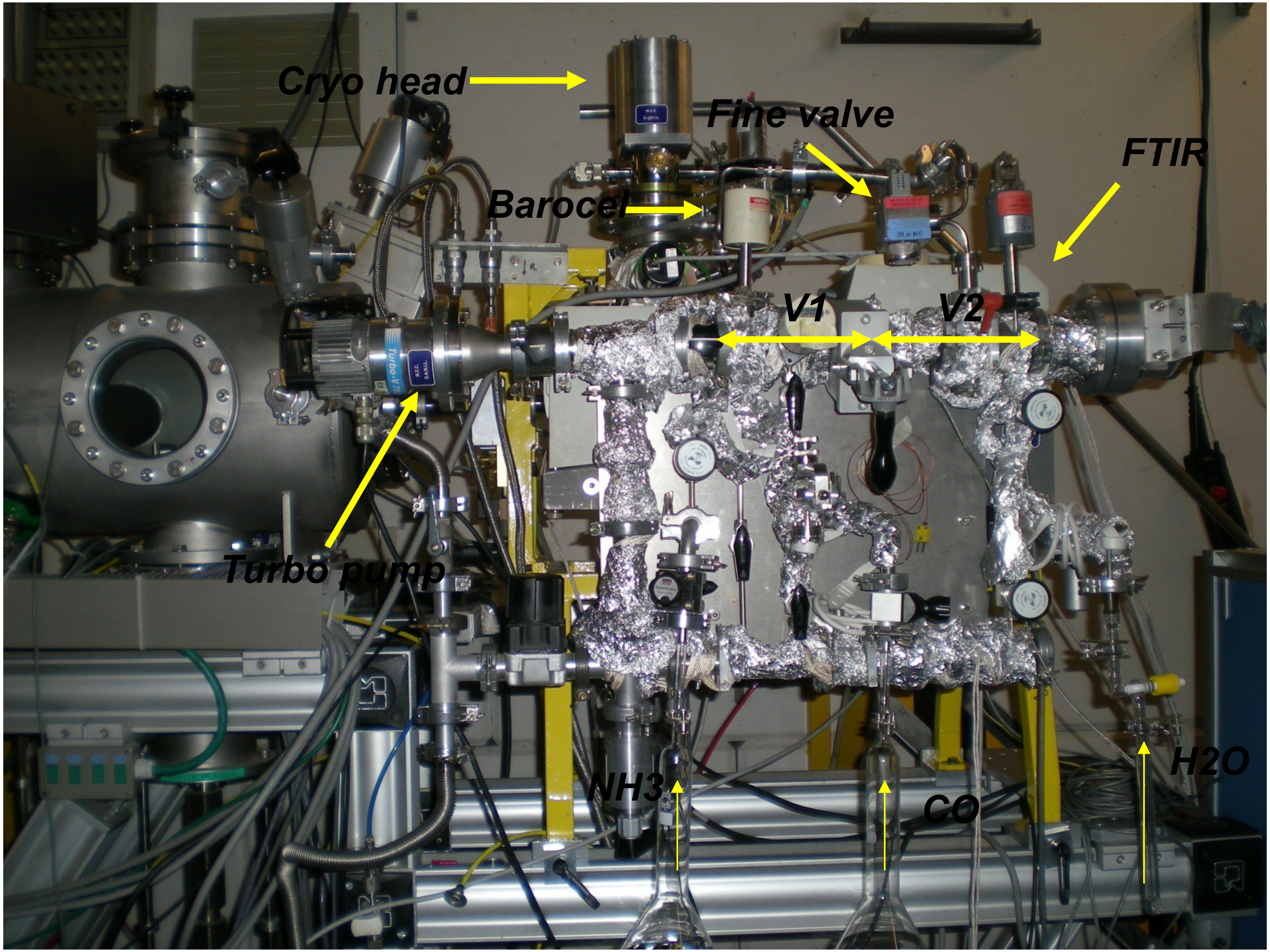
Se: 3 orders of magnitude

Cross sections versus Se

experimental set-up CASIMIR: FTIR of condensed gases at 14 K






Cryo head
Fine valve
FTIR
Barocel
V1
V2
Turbo pump
NH3
CO
H2O

Experimental details

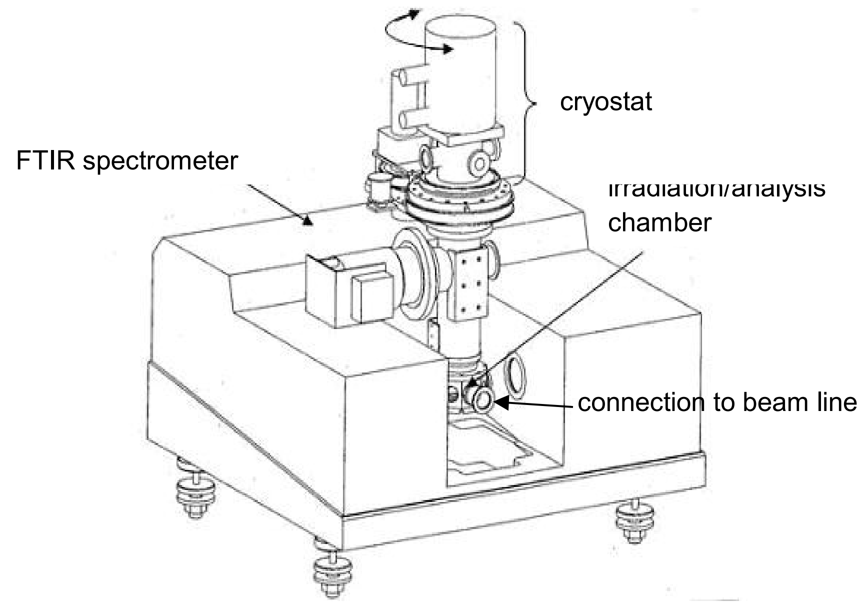
Pressure in irradiation chamber
~2x10-8 mbar (14 K)

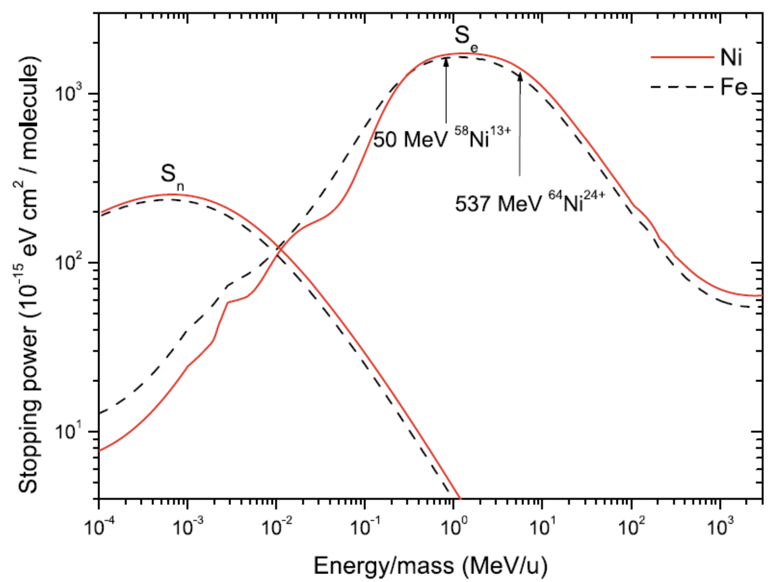
Substrate
CsI, ZnSe windows

Temperature
13 K < **T** < 300 K

Samples (ices)
- in situ gas deposition
- thickness ~0.1 - 2 μm (1017-1018 molecules/cm2)
- ion penetration depth > ice thickness (HE exp.)
- ion implantation (Low E exp.)

Ion beam (**G**rand **A**ccélérateur **N**ational d`**I**ons **L**ourds, Caen, France)
- 50 MeV 58Ni13+, 537 MeV 64Ni24+
- flux ~109 ion/cm2 s
- fluence upto 2x1013 ion/cm2 (typically 4 hours)





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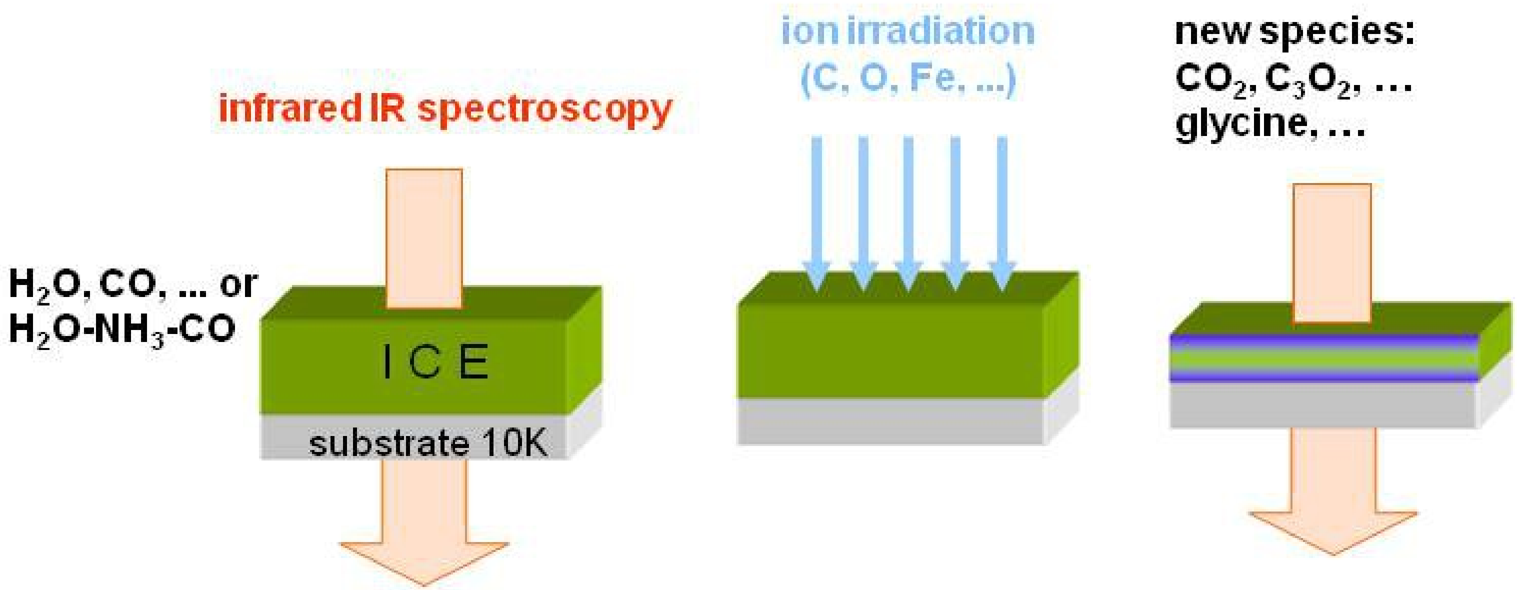
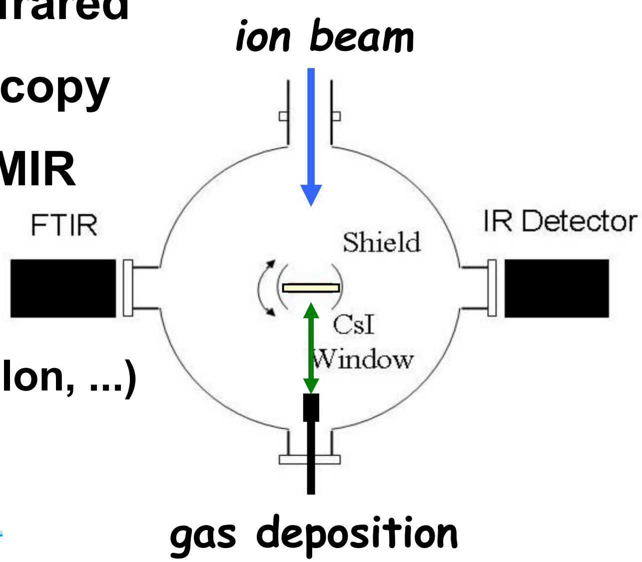
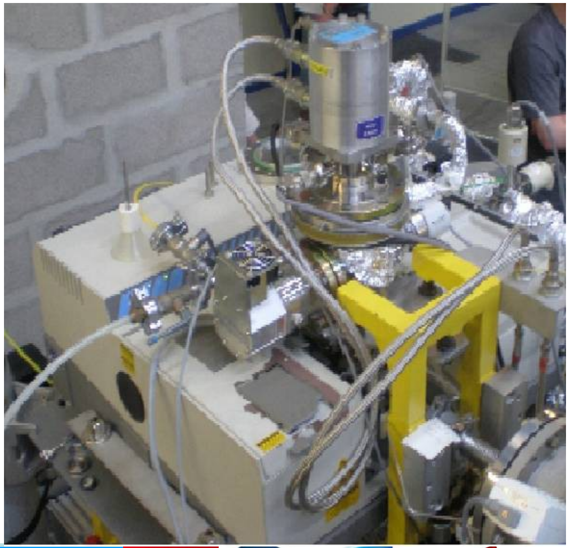


Fourier Transform Infrared Absorption Spectroscopy

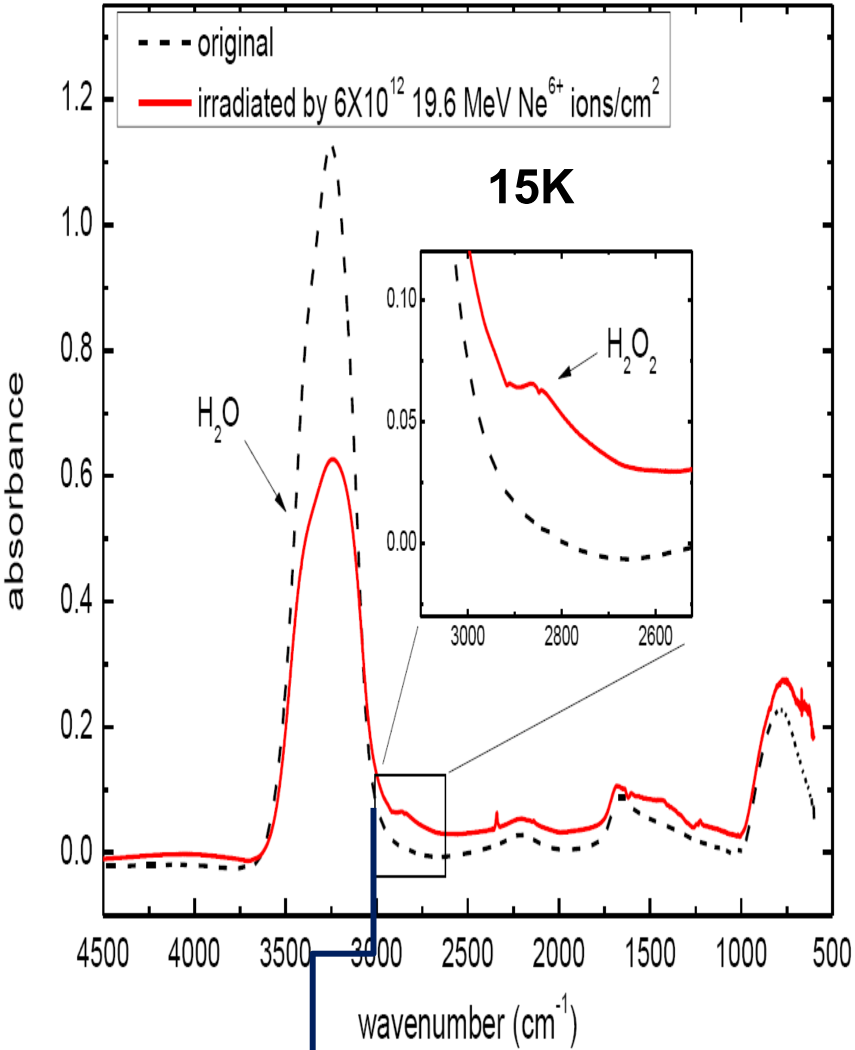
FTIR @CIMAP: CASIMIR





(E. Balanzat, J.M. Ramillon, ...)

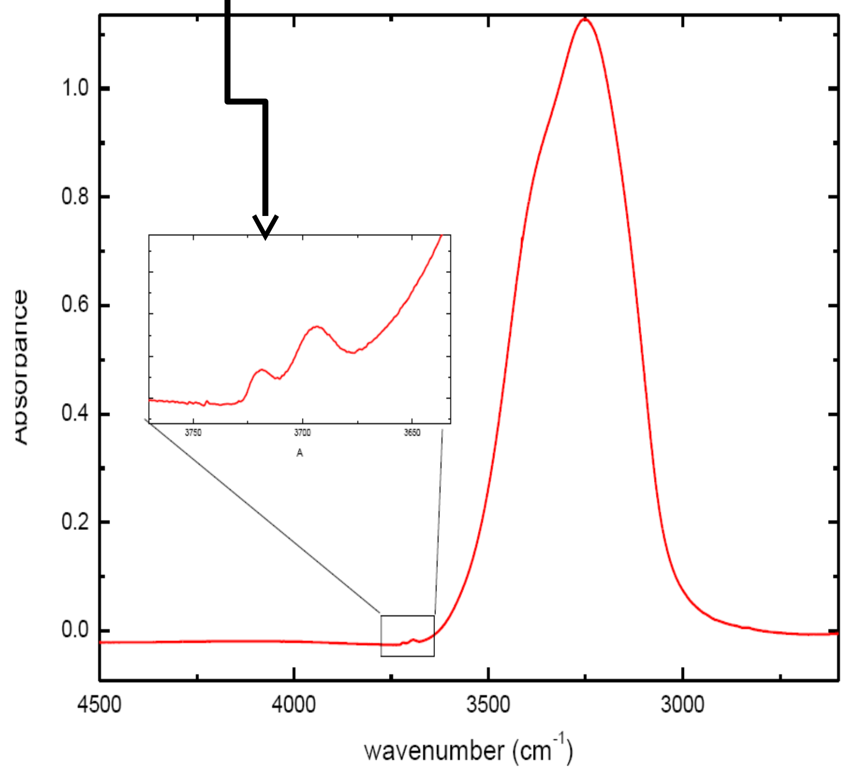
Water ice:
Compaction and Amorphization



The most abundant molecule in interstellar ices:

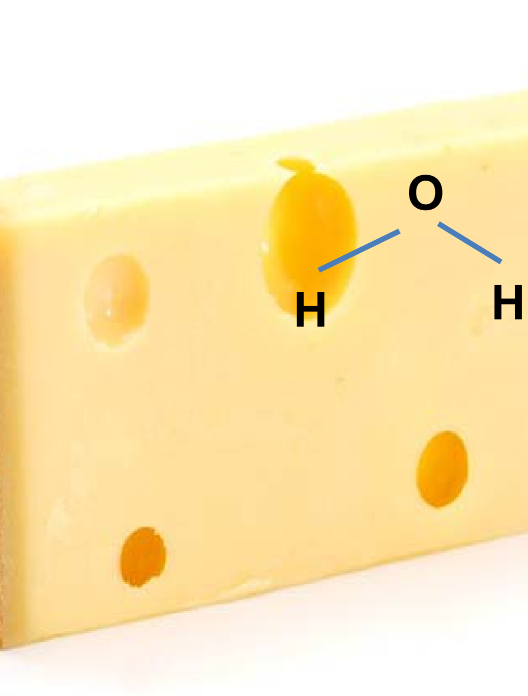
Water H2O

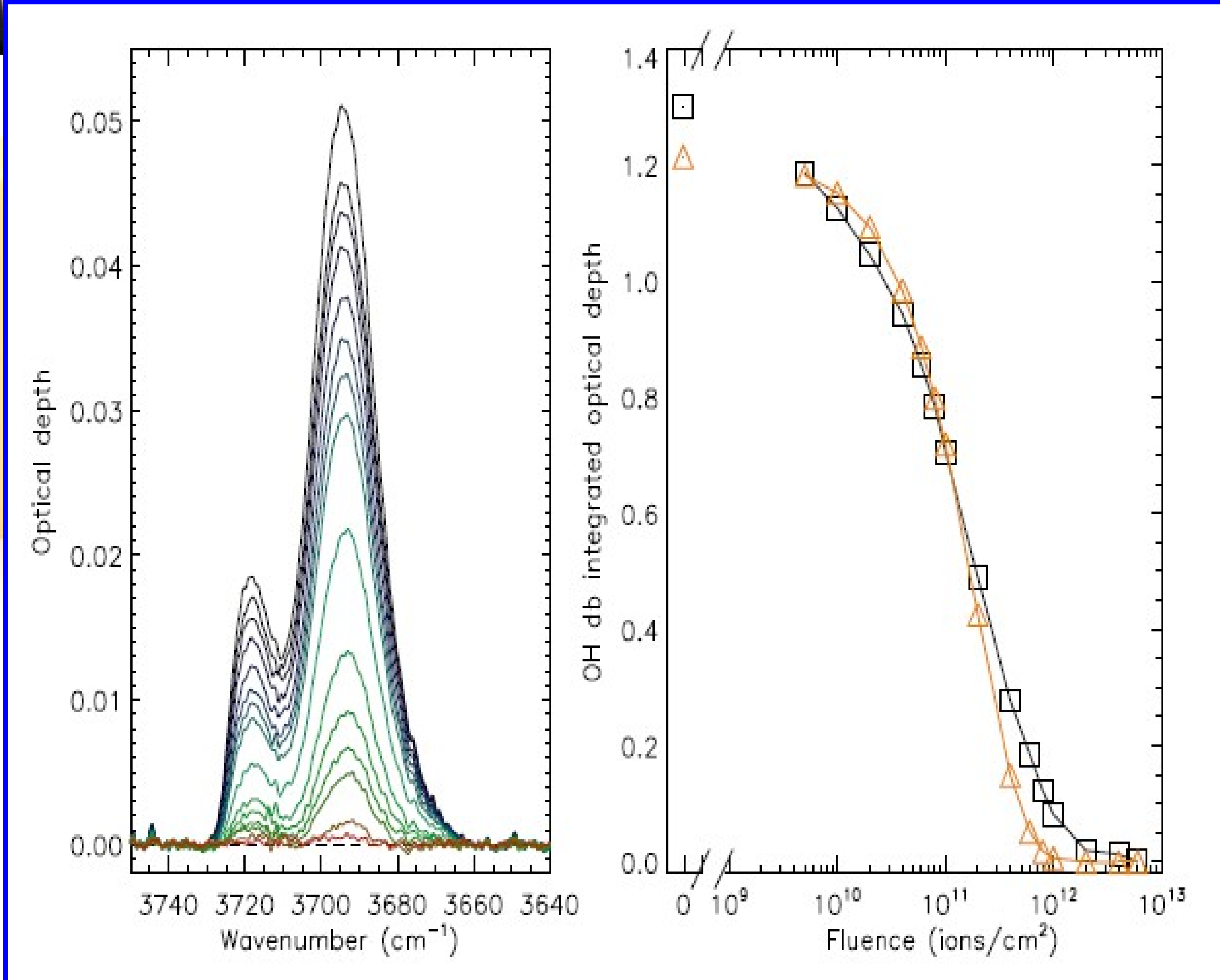
Porosity: OH dangling bonds



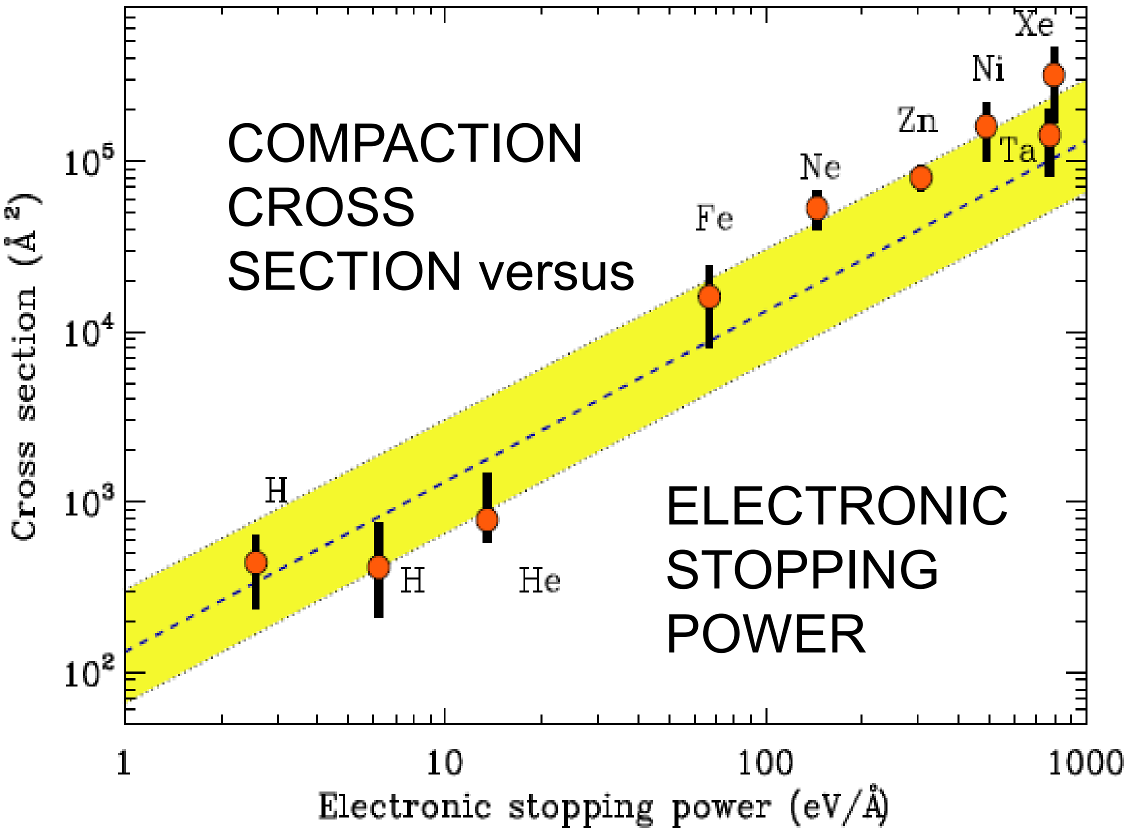
Irradiation of H2O ice: formation of H2O2

Centre de R





compaction "dose": 1 eV/molecule



σc = A Sen with n=1,0±0,2

tcomp = 1 x105 to 2 x106 years

small compared to cloud lifetimes

Indeed **no** OH dangling bonds observed by ISO in ISM

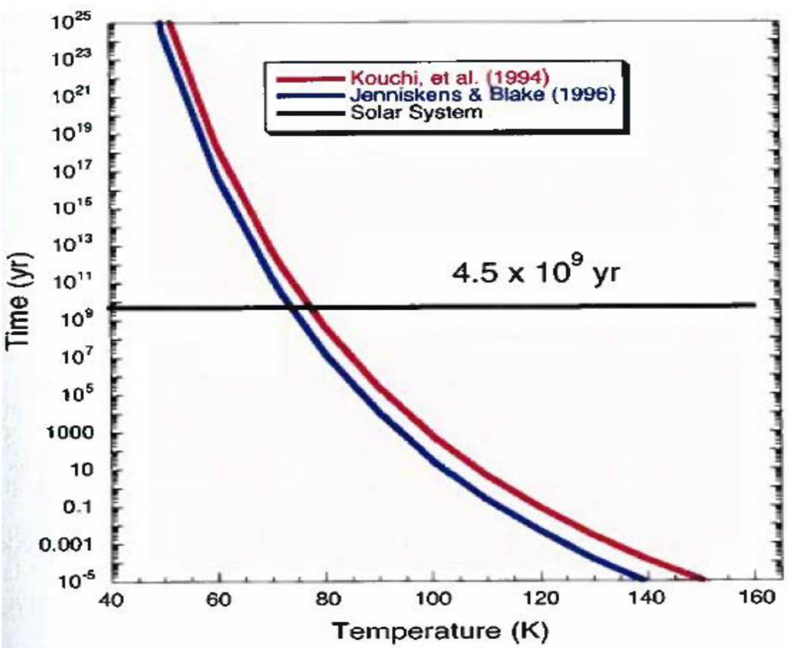
Compaction of Water Ice by Cosmic Rays: Experiment 2012 GANIL-LISE

E. Dartois, J.J. Ding, A.L.F. de Barros, P. Boduch, R. Brunetto, M. Chabot, A. Domaracka, M. Godard, X.Y. Lv, C.F. Mejia Guaman, T. Pino, H. Rothard, E.F. da Silveira, J.C. Thomas

Swift heavy ion irradiation of water ice at MeV to GeV energies: approaching true cosmic ray compaction

Astronomy & Astrophysics 557 (2013) A97

Crystal versus amorphous ice: a competition



Mastrapa et al, 2013

Thermal induced transition:

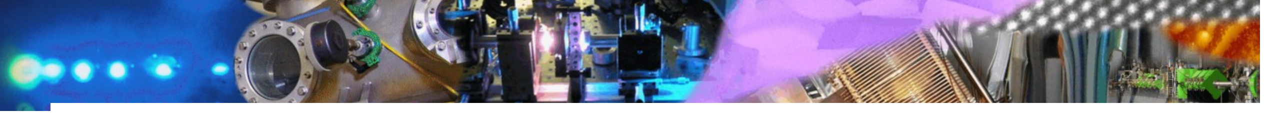
- At 100K amorphous ice converted in crystal in about 103 years.

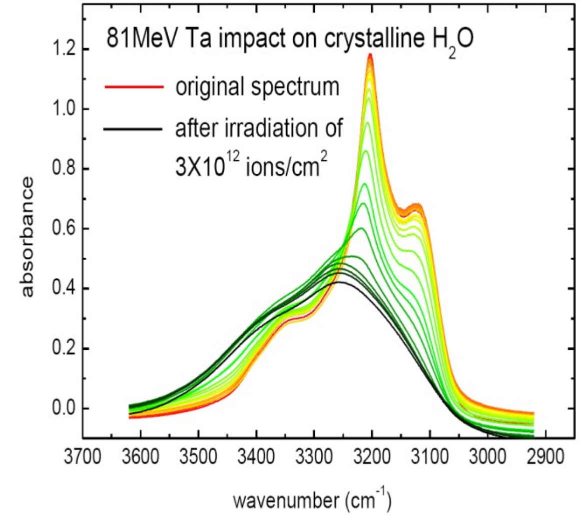
Irradiation : it induces amorphization.

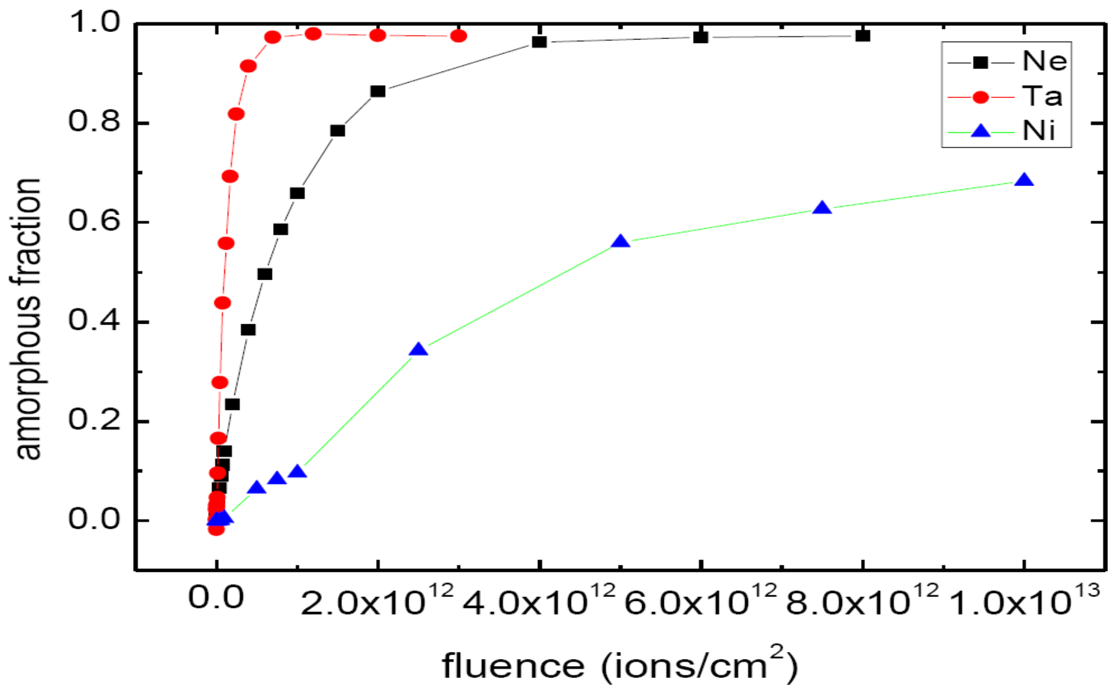
Table3-2: Ions used for irradiation, their electronic stopping power S_e, their nuclear stopping power S_n, and the irradiation temperature.

	Energy (MeV)	Irradiation temperature	S_e (eV/Å)	S_n (eV/Å)
Ne	19.6	15K	143	0.2
Ta	81	17K	757	12.7
Ni	46	145K	460	1.4

$$f = A_c \times f_c + A_a \times f_a$$







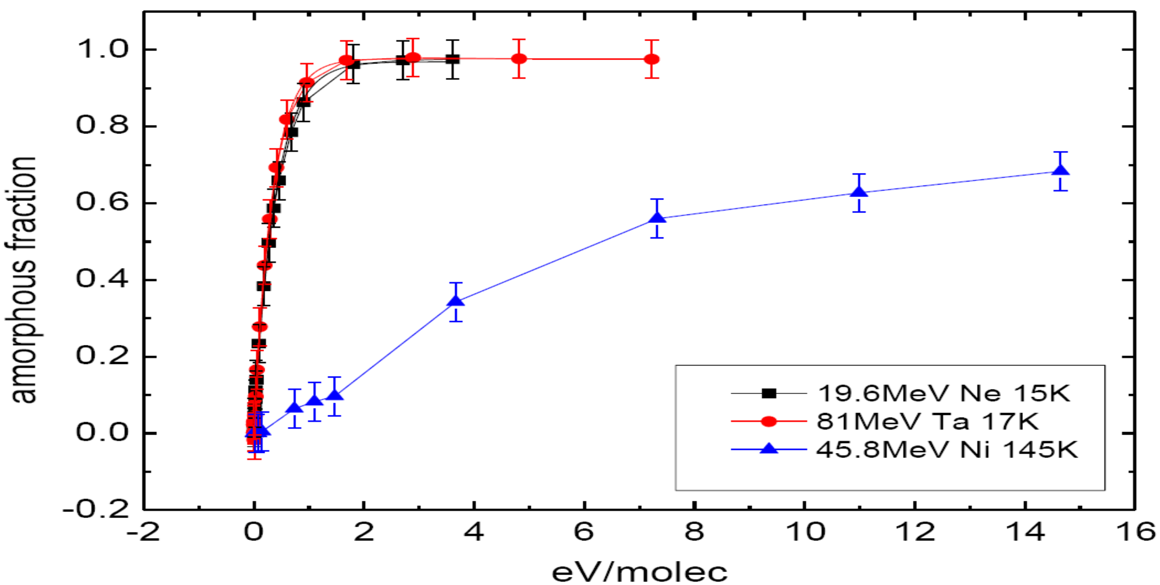
Local dose: the key parameter!

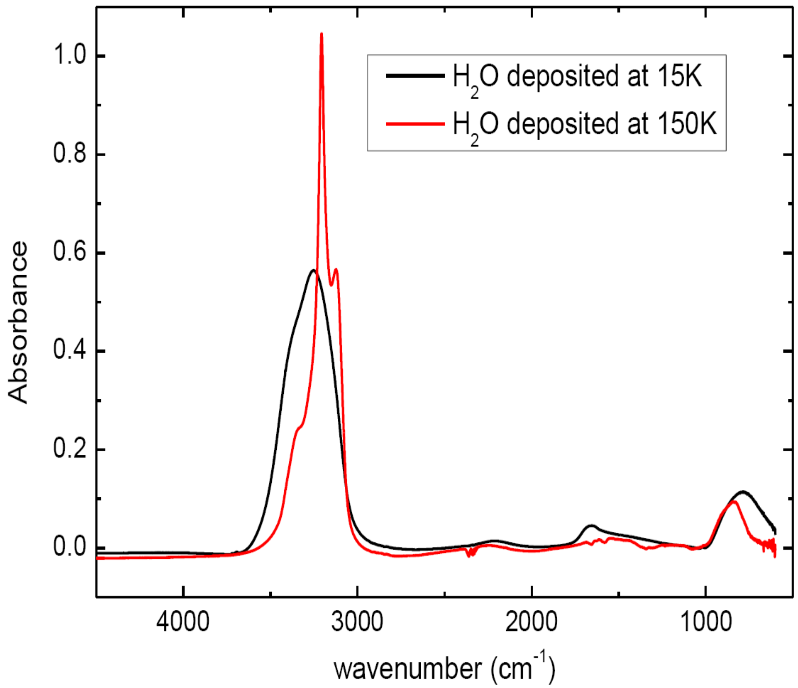
95% at 1,4 eV @15K

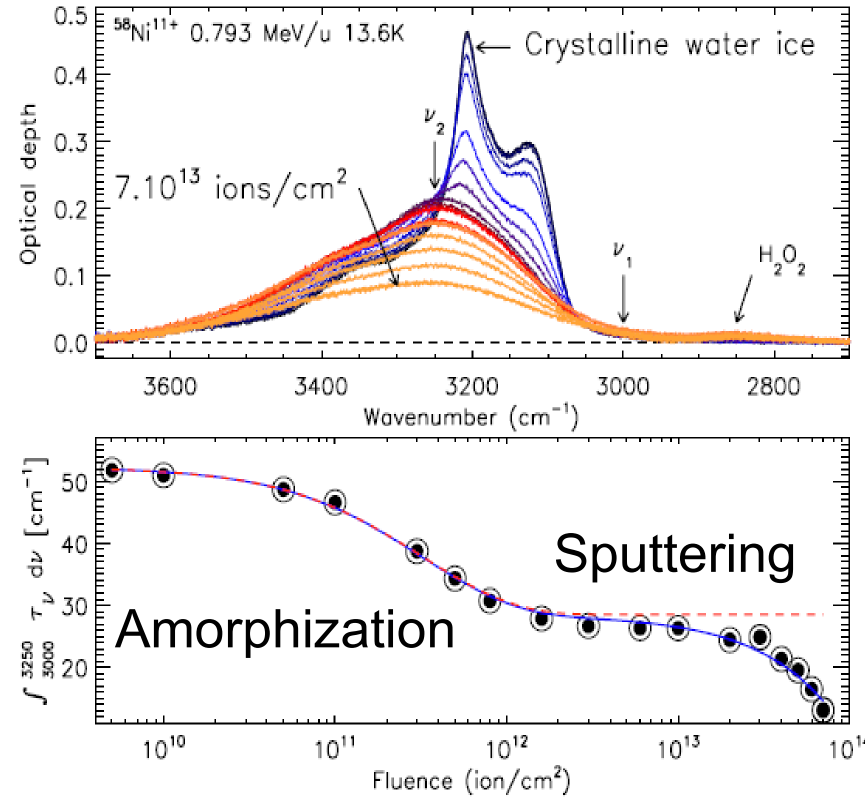
Around 3eV for 100%

At 145 K, much longer due to the thermal crystallization

Thesis JJ Ding







Total Amorphisation dose:
3 eV/molecule

Ion irradiation 3 times more efficient for compaction vs. amorphization
Water ice resilient to phase transition

End point:
amorphous compact ice

E. Dartois, B. Augé, P. Boduch, R. Brunetto, M. Chabot, A. Domaracka, J.J. Ding, O. Kamalou, X.Y .Lv, H. Rothard, E.F. da Silveira, J.C. Thomas
Heavy ion irradiation of crystalline water ice -Cosmic ray amorphization cross-section and sputtering yield
Astronomy & Astrophysics 576 (2015) A126

Carbon Oxide CO,

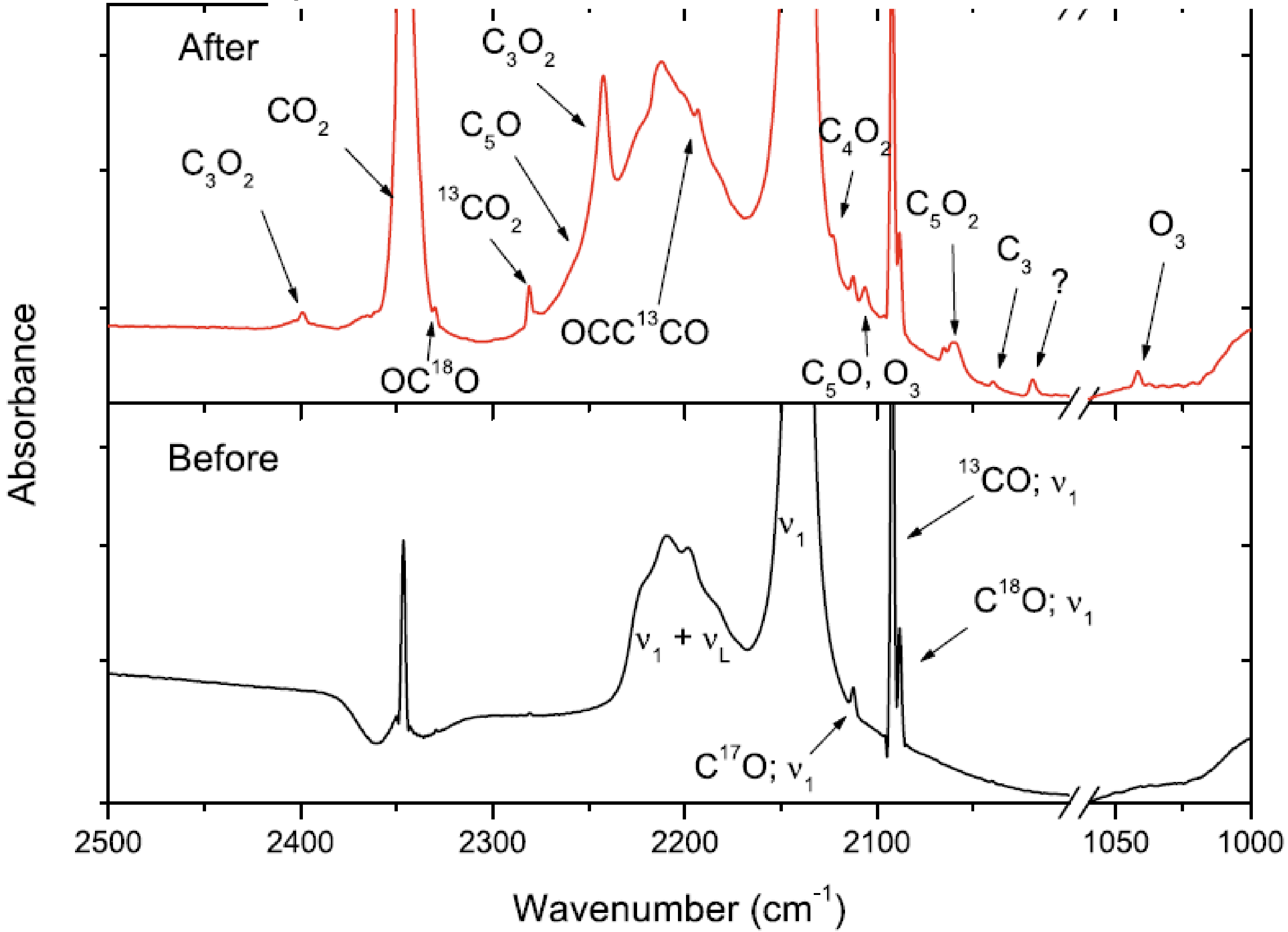
dense molecular clouds,

and cosmic rays.

The starting point: the Eduardo's thesis, cotutella with Enio.

Second example: CO ice

(the second most abundant molecule in space ices after H2O)



CO destruction

Production of CO2, O3, Cx and CxOy

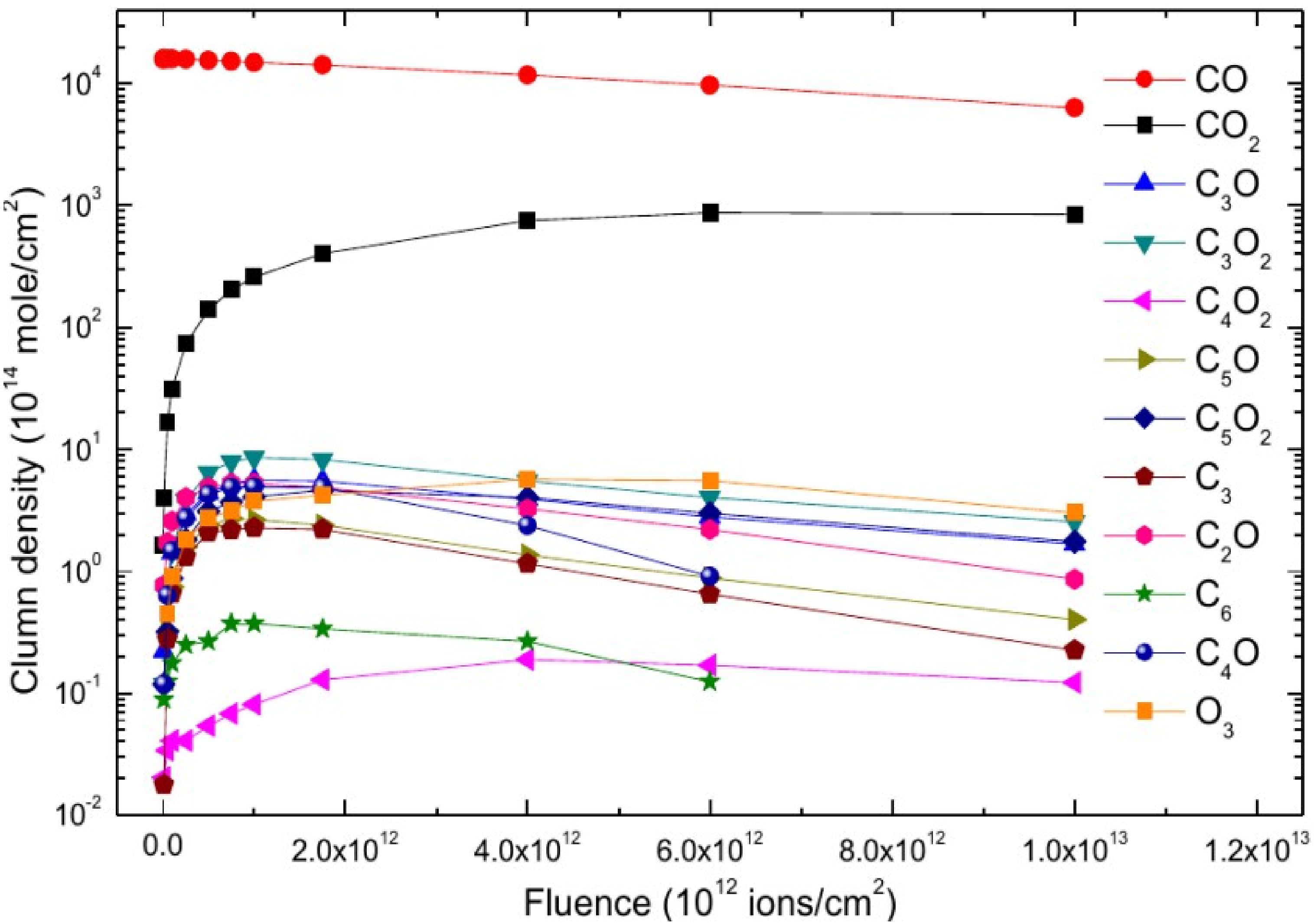
Main « observable » daugther: CO2

Infrared spectrum of CO ice before and after 50 MeV $^{58}Ni^{11+}$ irradiation with a fluence of 1.0×10^{12} cm^{-2}.

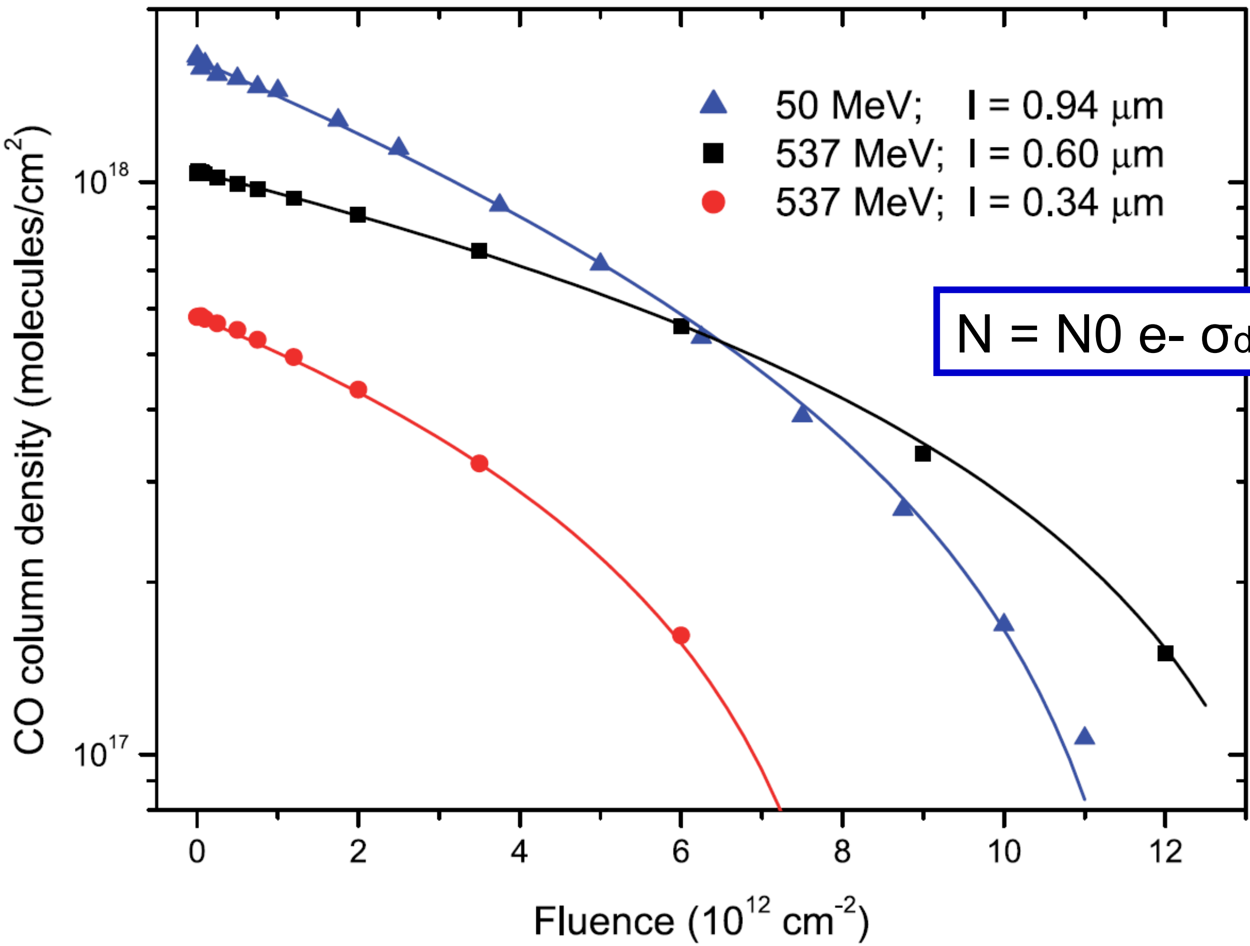
ion	E_0	S_e	S_n	P_d	ℓ_0	N_0
$^{16}O^{7+}$	220	94	0.04	812	0.41	7.16
$^{16}O^{5+}$	16	385	0.4	25	0.39	6.78
$^{16}O^{2+}$	6	452	1.0	11	0.53	9.22
$^{64}Ni^{24+}$	537	1136	0.7	226	0.39	6.88
$^{70}Zn^{26+}$	606	1255	0.7	228	0.74	12.86
$^{56}Fe^{24+}$	270	1318	1.0	112	0.24	4.15
$^{58}Ni^{11+}$	46	1690	5.5	29	0.85	14.8
$^{58}Ni^{13+}$	52	1706	4.9	31	0.54	9.45
$^{58}Ni^{13+}$	52	1706	4.9	31	0.66	11.5
$^{86}Kr^{31+}$	774	1731	1.1	233	0.05	0.83

Sputtering yield , destruction and formation cross sections…
… as a function of Se, the electronic stopping power

CO ice: formation of new molecular species



CO ice: disappearence of CO Molecules during Nickel Ion Irradiation:



$N = N0\ e^{-\sigma_d F} - (\mathbf{Y}/\sigma_d)(1 - e^{-\sigma_d F})$

deduced quantities:

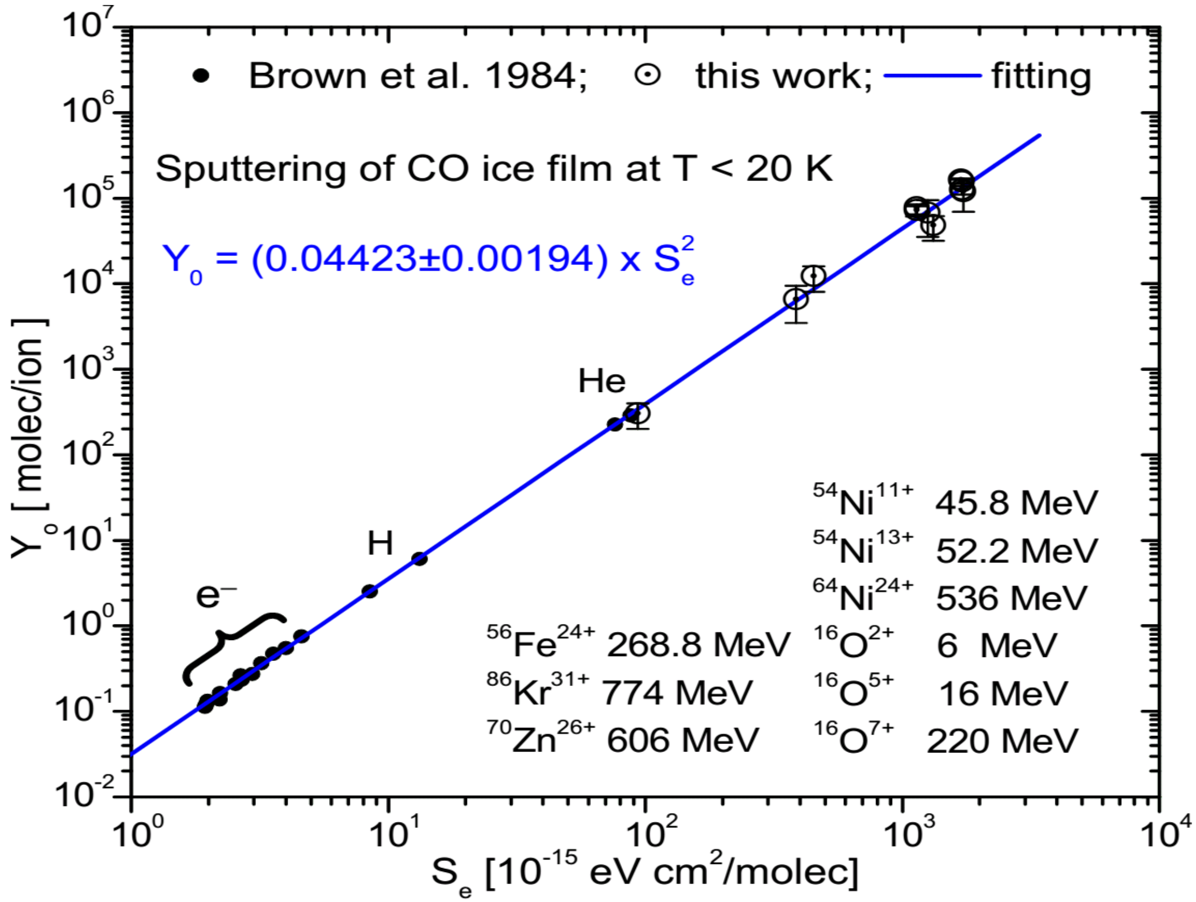
Destruction Cross Section σ_d

Sputtering Yield Y

Centre de Recherche sur les Ions, les Matériaux et la Photonique

nom

CO ice: Ion induced Sputtering Yield



Y~Se

Se ~ ZP2**2**

Y ~ Se2

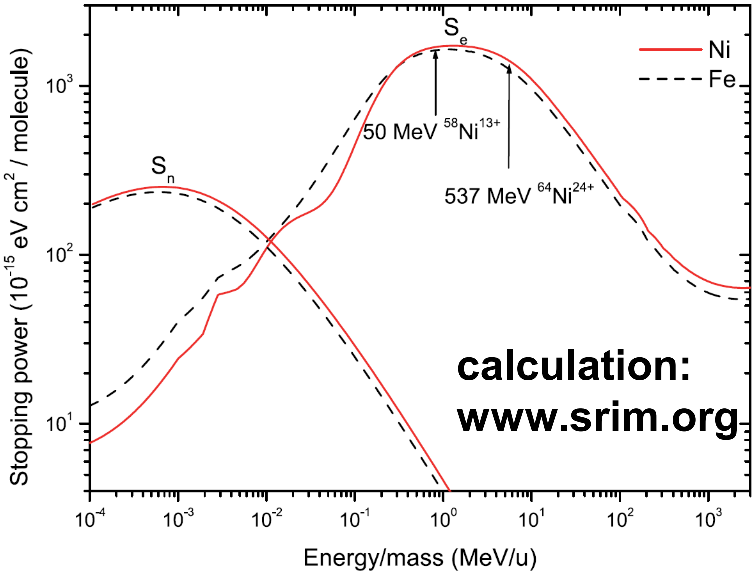
Y ~ ZP4

very strong dependence!

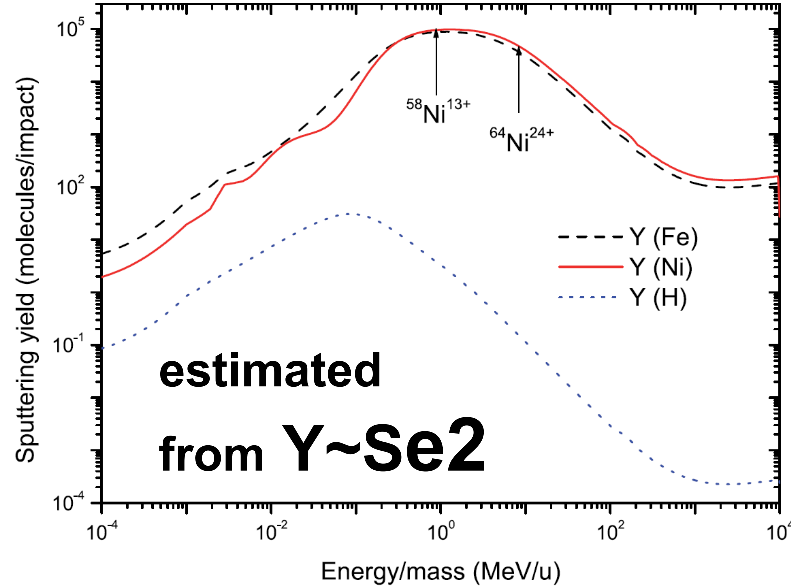
W.L. Brown, W.M. Augustyniak, K.J. Marcantonio, E.H. Simmons, J.W. Boring, R.E. Johnson, C.T. Reimann, Nucl. Instrum. Meth. B1 (1984) 307

E. Seperuelo Duarte, A. Domaracka, P. Boduch, H. Rothard, E. Dartois, E.F. da Silveira Astronomy & Astrophysics 512 (2010) A71

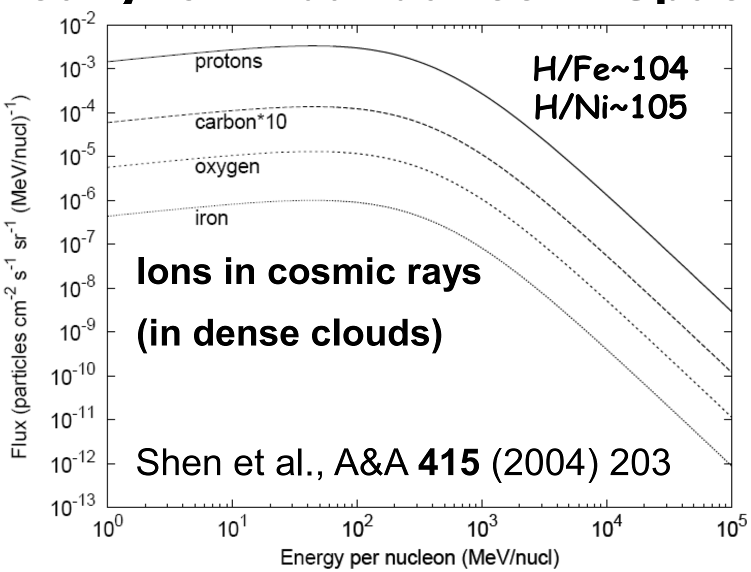
Electronic Energy Loss in CO



CO Sputtering Yield



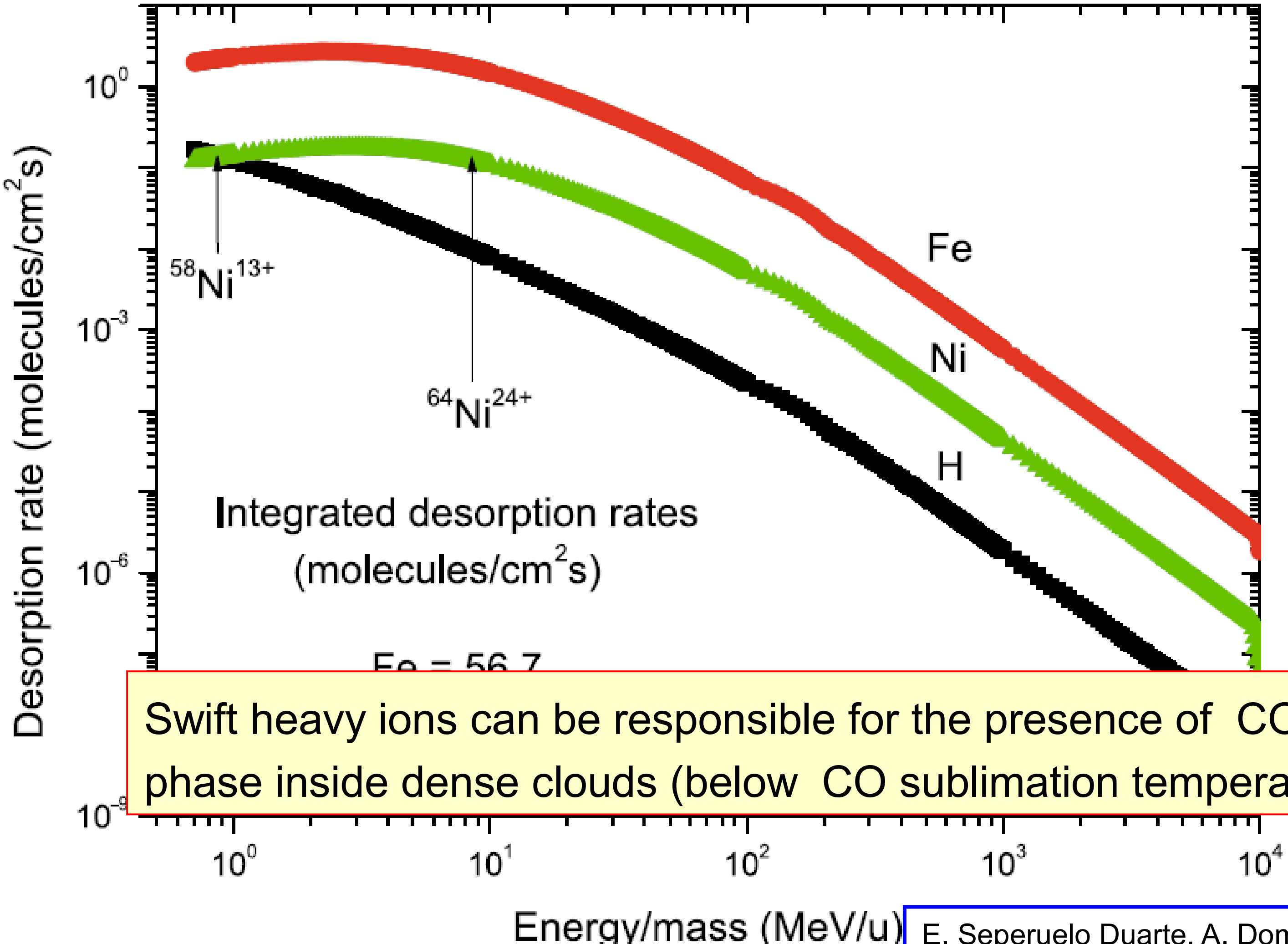
Heavy Ion Abundance in Space



astrophysical application:

presence of CO in the gas phase in "dense" (104–106 molecules cm-3) molecular clouds ?

Astrophysical implication

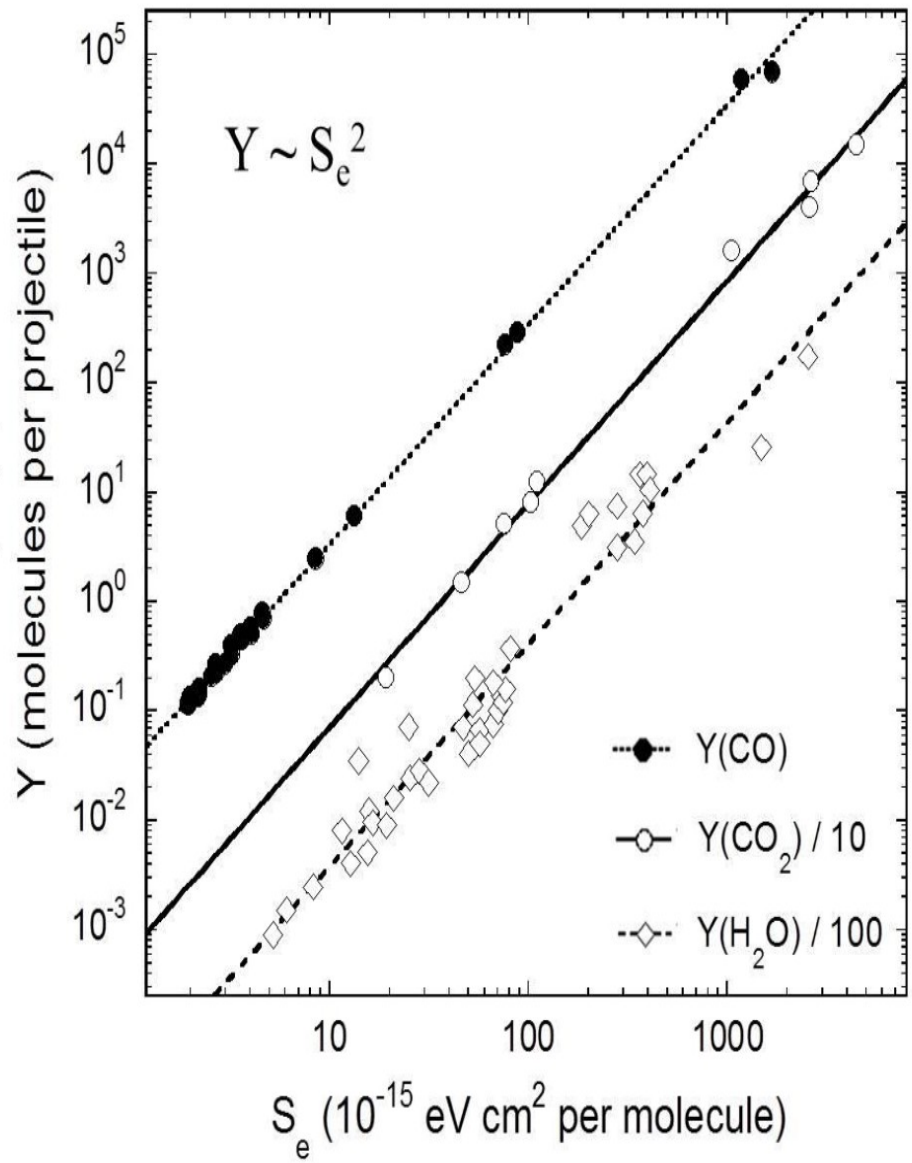


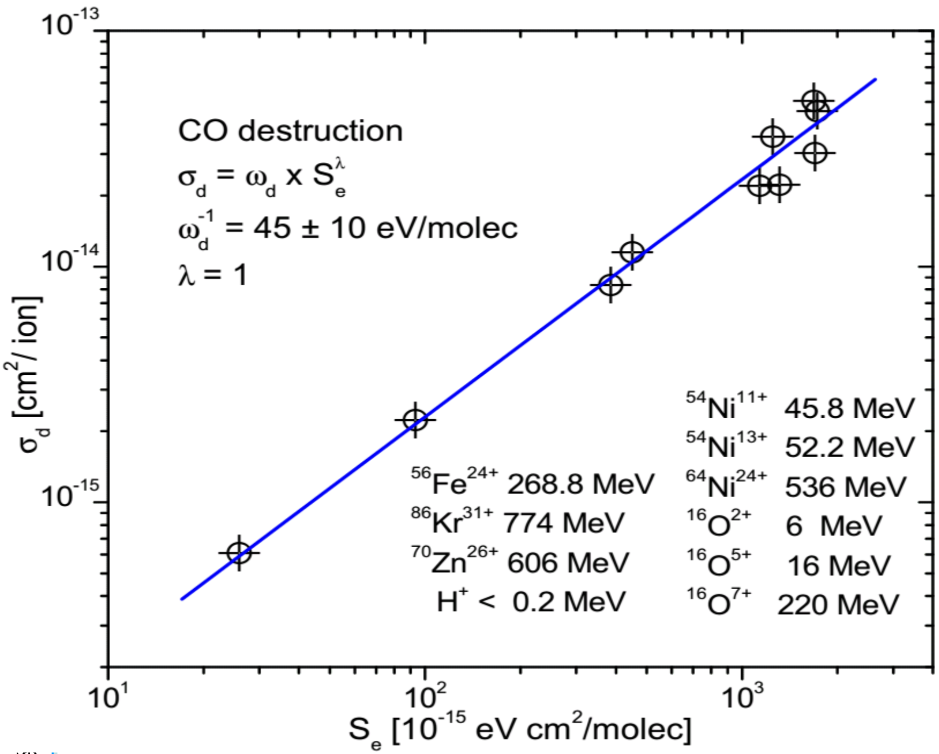
Swift heavy ions can be responsible for the presence of CO in the gas phase inside dense clouds (below CO sublimation temperature,10 K).

E. Seperuelo Duarte, A. Domaracka, P. Boduch, H. Rothard,E. Dartois, E.F. da Silveira
Laboratory simulation of heavy ion cosmic ray interaction with condensed CO
Astronomy & Astrophysics 512 (2010) A71

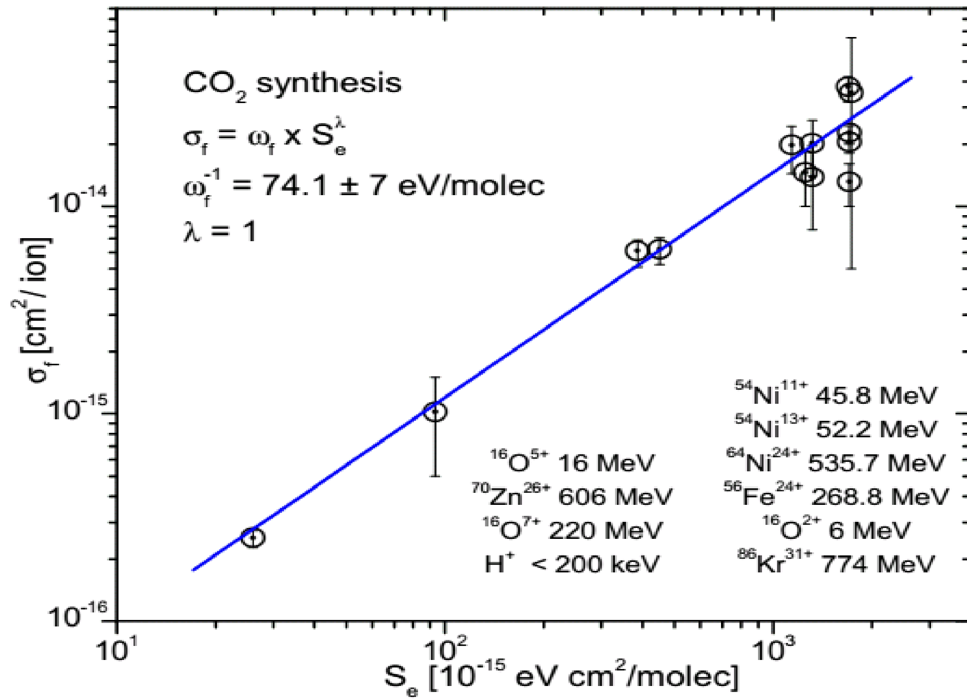
The same results for:

CO, CO2 and H2O





$\sigma_{d\,(CO)}\ \alpha\ Se$



$\sigma_{f\,(CO2)}\ \alpha\ Se$

Formation proportional to destruction :
Chemical Equilibrium ?

Destruction and formation

CO ice-different projectiles: destruction/formation cross sections

Comparison with "other projectiles"

Molecules	Projectile	σ (10^{-15} cm^2)	Reference
CO destruction	50 MeV Ni^{13+}	100	This work
	537 MeV Ni^{24+}	30	This work
	200 keV H^+	0.28	Loeffler et al. (2005)
	10.2 eV photons	0.0003	Loeffler et al. (2005)
	>6 eV photons	<0.000001	Cottin et al. (2003)
	>6 eV photons	<0.00008	Gerakines et al. (1996)
CO_2 formation	50 MeV Ni^{13+}	20	This work
	537 MeV Ni^{24+}	18	This work
	200 keV H^+	6	Loeffler et al. (2005)
	10.2 eV photons	0.017	Loeffler et al. (2005)
	>6 eV photons	0.000013	Gerakines et al. (1996)

Astron. Astrophys. **512** (2010) A71

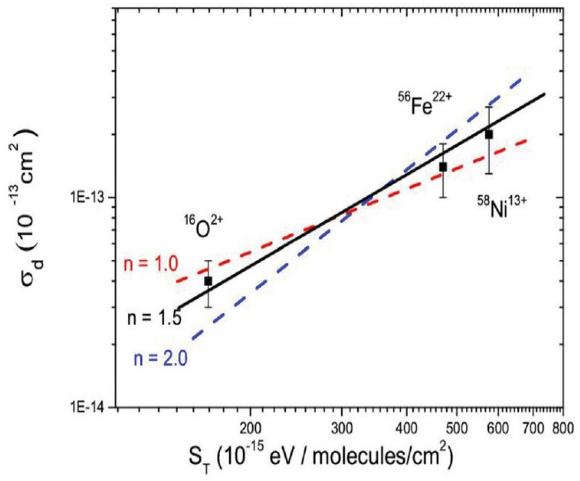


Figure 6. The dependence of the HCOOH destruction cross-section on the total stopping power. Data are for 6 MeV (O), 52 MeV (Ni; in preparation) and 267 MeV (Fe; the results of the current work). The lines correspond to the function $\sigma_d \sim S_e^n$, for $n = 3/2$ (solid line).

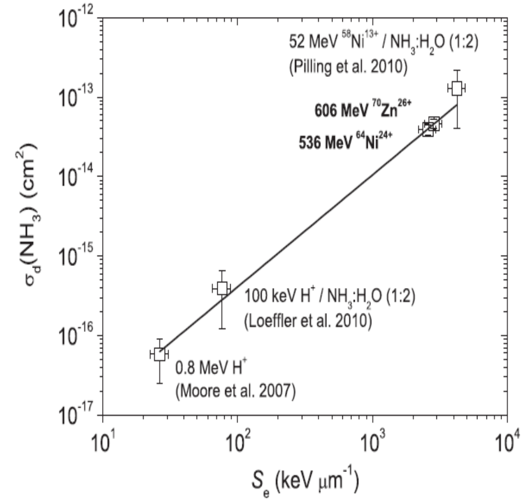


Figure 16. Destruction cross-section (σ_d) and stopping power (S_e) relationship The power law $\sigma_d(NH_3) \propto S_e^{1.4\pm0.1}$ is derived from $\sigma_d(NH_3)$ obtained in thi work and those compiled from the literature. See details in the text.

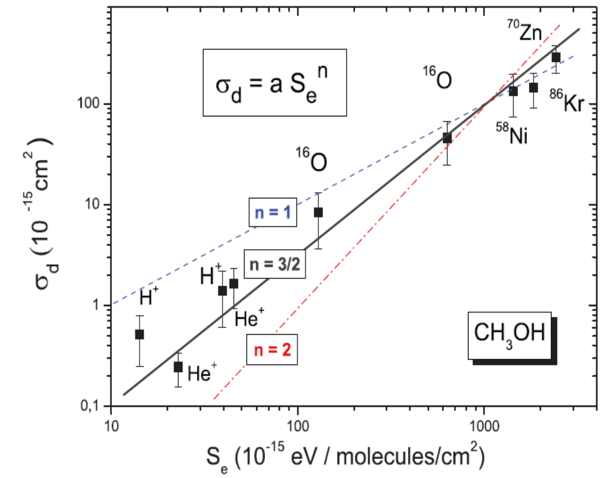


Figure 8. The dependence of CH_3OH destruction cross-section on the electronic stopping power. Data for 16- and 220-MeV O, Zn and Kr are results of the current work; Gerakines et al.(2001), Brunetto et al. (2005) and Baratta et al.(2002). The lines correspond to the function $\sigma_d \sim S_e^n$, for $n = 1$, 3/2 (solid line) and 2.

Diana P. P. Andrade et al(MNRAS 2013)

Vinicius Bordalo et al (Astro. Journal (2013)

Ana L, F, de Barros et al (MNRAS 2011)

n=1,5 for formic acid

n=1,4 for ammonia

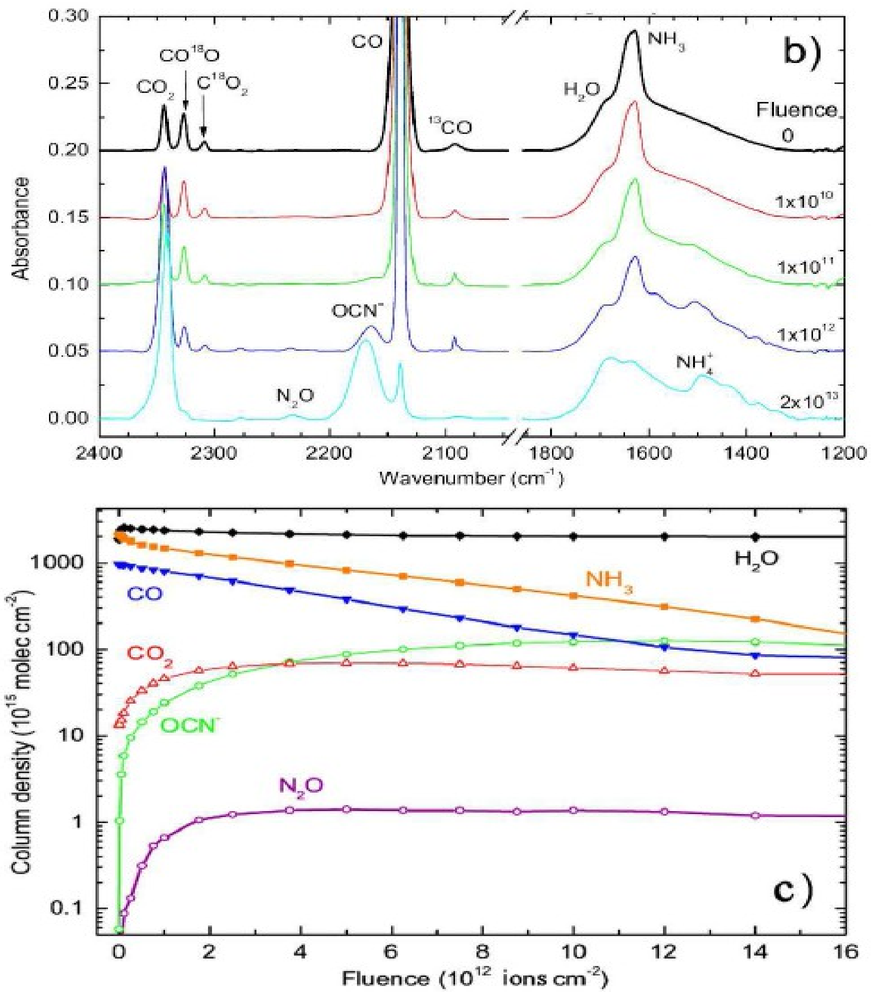
n=1.5 for methanol

Conclusion: for the destruction, always between 1 and 1,5 for simple molecules

Mixtures and complex organic molecules

H2O – CO - NH3 ice

46 Mev 58Ni13+



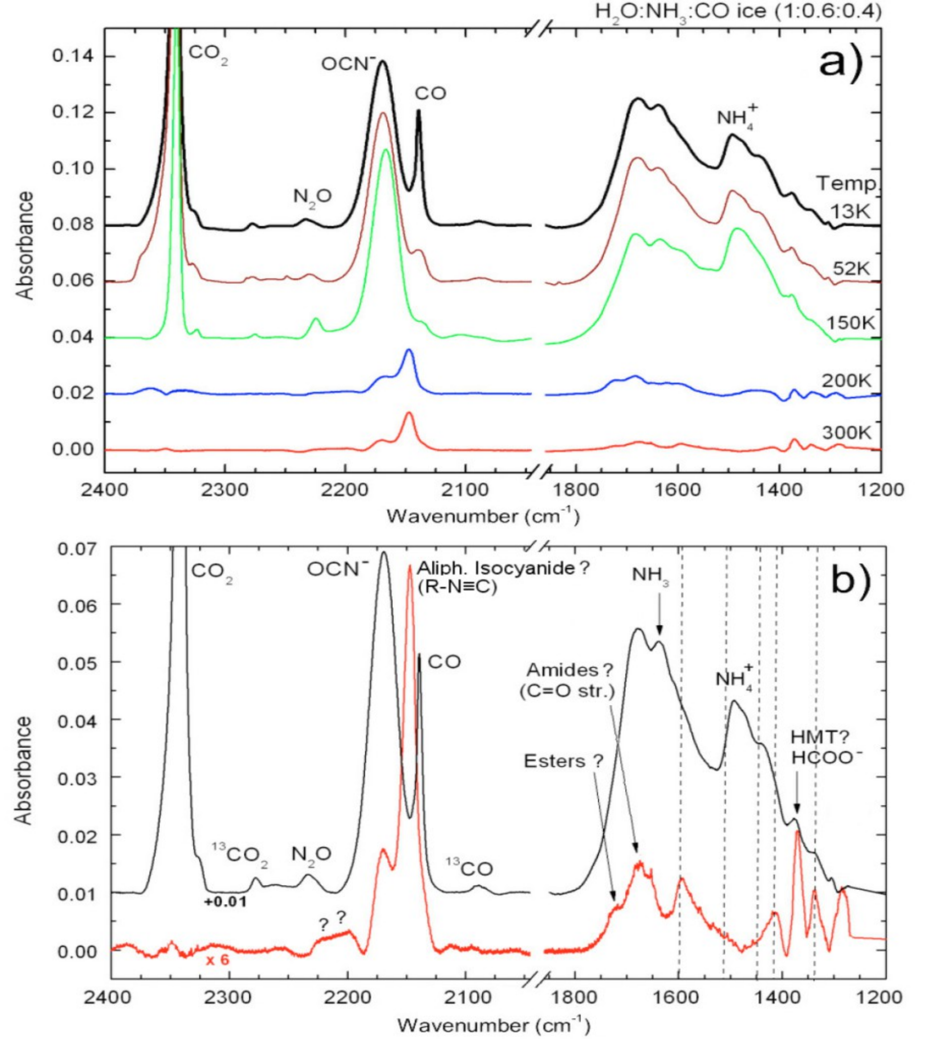


Fig. 6. a) Infrared spectra of $H_2O:NH_3:CO$ ice (1:0.6:0.4) from 2400 to 1200 cm^{-1} during heating to room temperature. The sample temperature of each spectrum is given. Each spectrum has an offset of 0.02 for clearer visualization. **b)** Comparison between the irradiated ice at 13 K (top spectrum) and the 300 K residue (bottom spectrum). Vertical dashed lines indicate the frequencies of some vibration modes of zwitterionic glycine ($NH_3^+CH_2COO^-$).

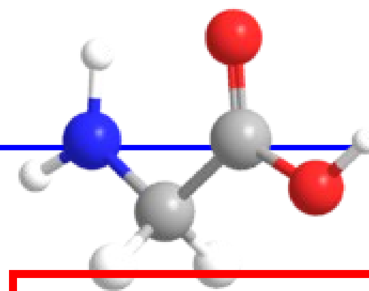
S. Pilling et al.Astronomy & Astrophysics 509 (2010) A87

Frequency (cm^{-1})	Wavelength (μm)	Temp. (K)	Molecule
2233	4.48	13	N_2O
2218–2200	4.51-4.54	300	nitriles†
2168	4.61	13, 300	OCN^-
2147	4.66	300	aliph. isocyanide†
~2112	4.73	300	$NCO_2^{\dagger}$
1725	5.80	300	ester†
1683	5.94	300	amides†
1652	6.05	300	asym-$N_2O_3^{\dagger}$
1637	6.11	13	?
1593	6.28	300	$NH_3^+CH_2COO^{-\dagger}$
1558	6.42	300	?
1533	6.52	300	?
1506	6.64	300	$NH_3^+CH_2COO^{-\dagger}$
~1490	6.71	13	NH_4^+
1474	6.78	13	NO_3^{-}
1440	6.94	13	$NH_3^+CH_2COO^{-\dagger}$
1415	7.07	300	$NH_3^+CH_2COO^{-\dagger}$
~1370	7.30	13, 300	HMT† $HCOO^-$
~1338	7.47	13, 300	$NH_3^+CH_2COO^{-\dagger}$ $NH_2CH_2COO^{-\dagger}$ $HCOO^-$
1305	7.66	13	$N_2O_3^{\dagger}$; $N_2O_4^{\dagger}$
1283	7.80	300	$N_2O^{\dagger}$

S. Pilling, E. Seperuelo Duarte, E. F. da Silveira, E. Balanzat, H. Rothard, A. Domaracka, P. Boduch
Radiolysis of ammonia-containing ices by energetic, heavy and highly charged ions inside dense astrophysical environments, Astronomy & Astrophysics 509 (2010) A87

H_2O - CO - NH_3 ice

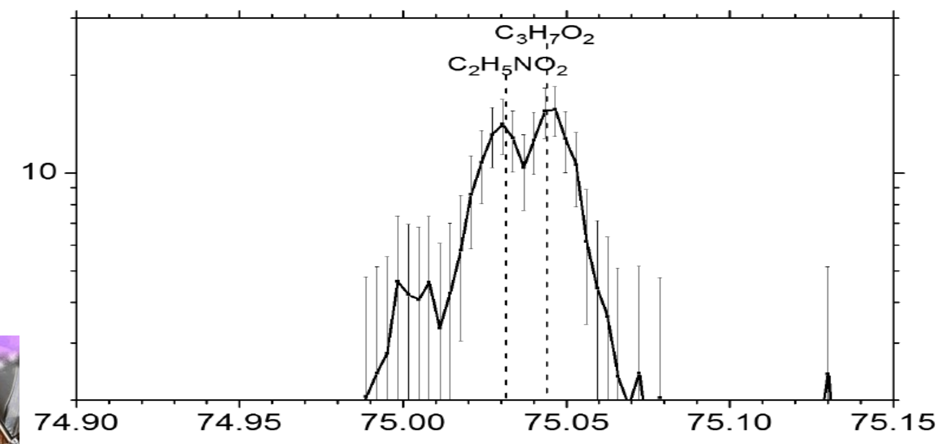
⇨ **glycine (amino acid)**



Analysis of the Residues by Chromatography?

The amount of residue?

Prebiotic chemicals
amino acid in the coma of
comet 67P/Churyumov-Gerasimenko



Kathrin Altwegg et al, Space sciences, 2016.

New experimental setup : IGLIAS

- 1 10-10 mbar (1 ML of water per hour)
- Online device with two spectrometers:
  - IR Bruker V70 (under primary vaccum, (500-6000 cm-1)
  - UV visible Perkin (200-800 nm, transmission, optical fiber).
  - for samples: 3 windows, 20 mm diameter (bigger residues).
  - Up to 4 gas for the deposition, co deposition avalaible.
  - QMS, electron gun.

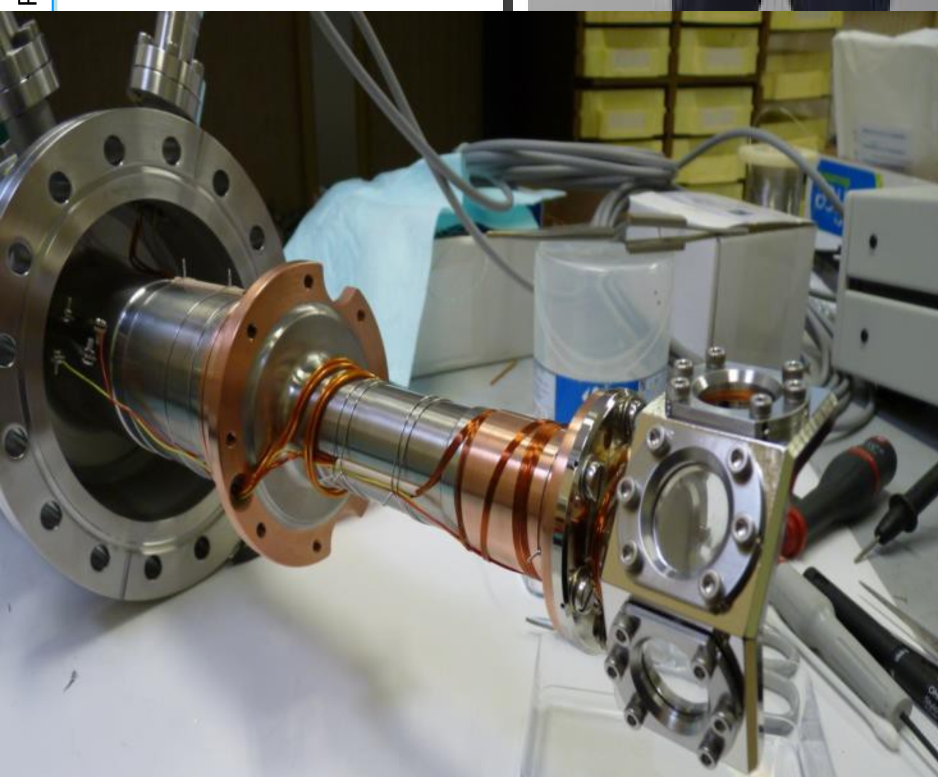
- Open to the scientific communitee!

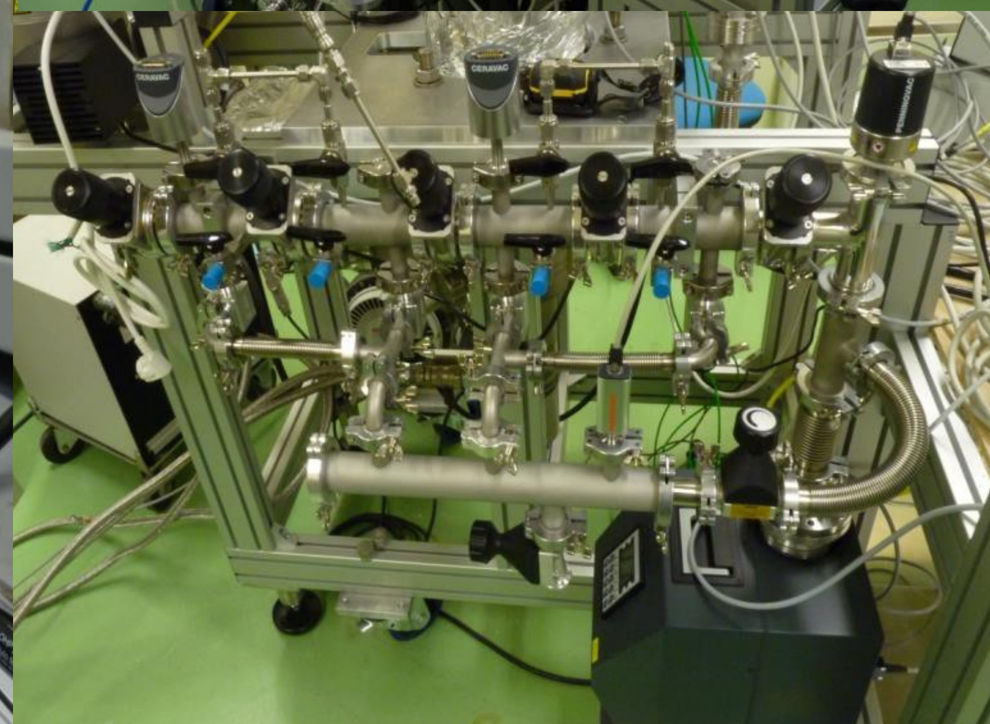
ANR IGLIAS

Ph. Boduch
E. Dartois

B. Augé (thesis)

Photonique





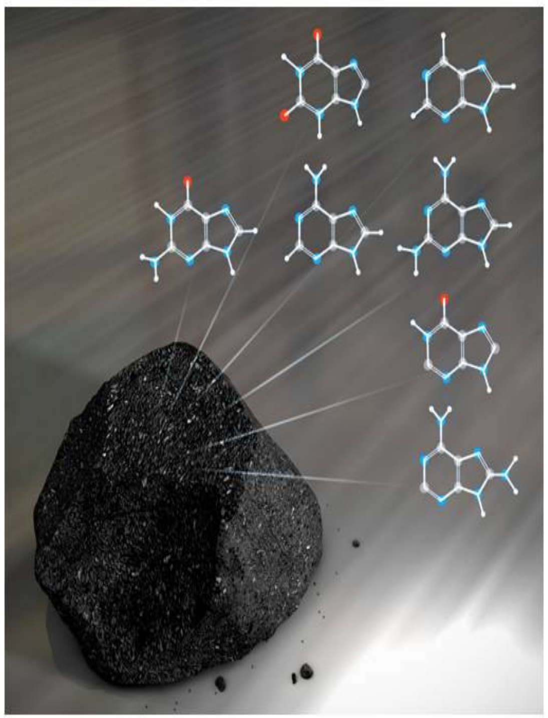
Destruction of COM:

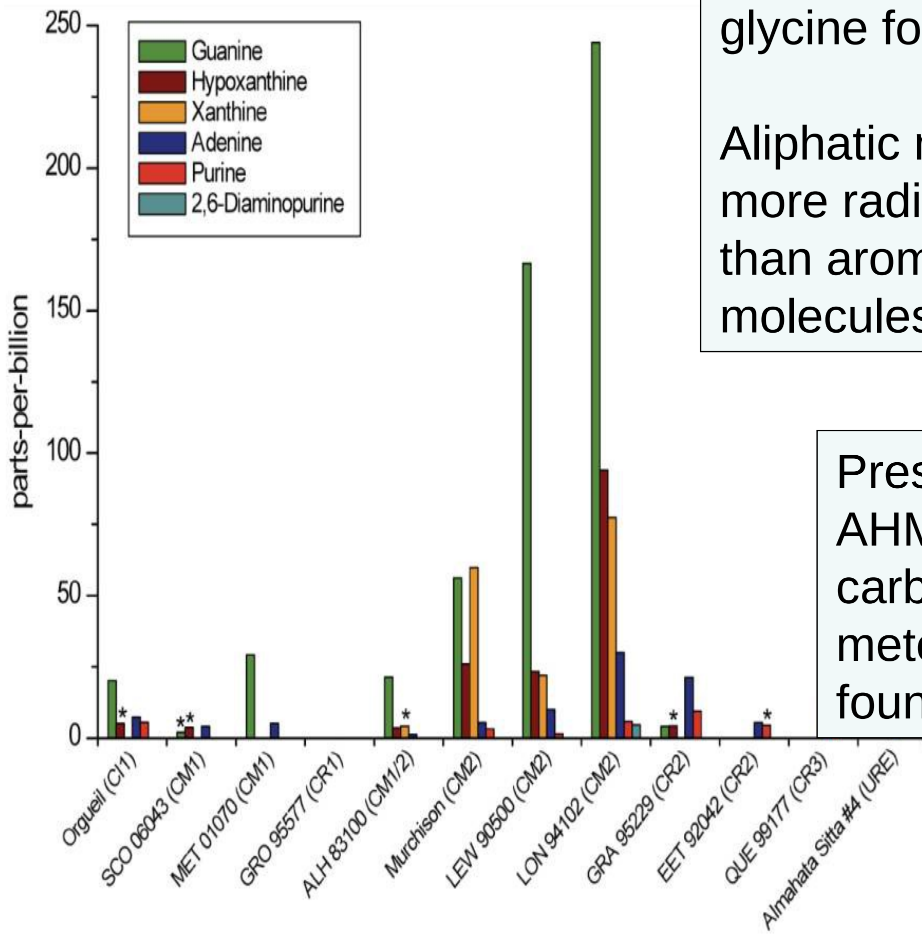
Irradiation of nucleobases

Introduction – Do AHMs exist in space?

AHMs have not yet been detected

Strong evidences:





Complex organic molecules already were detected in outer space, glycine for example.

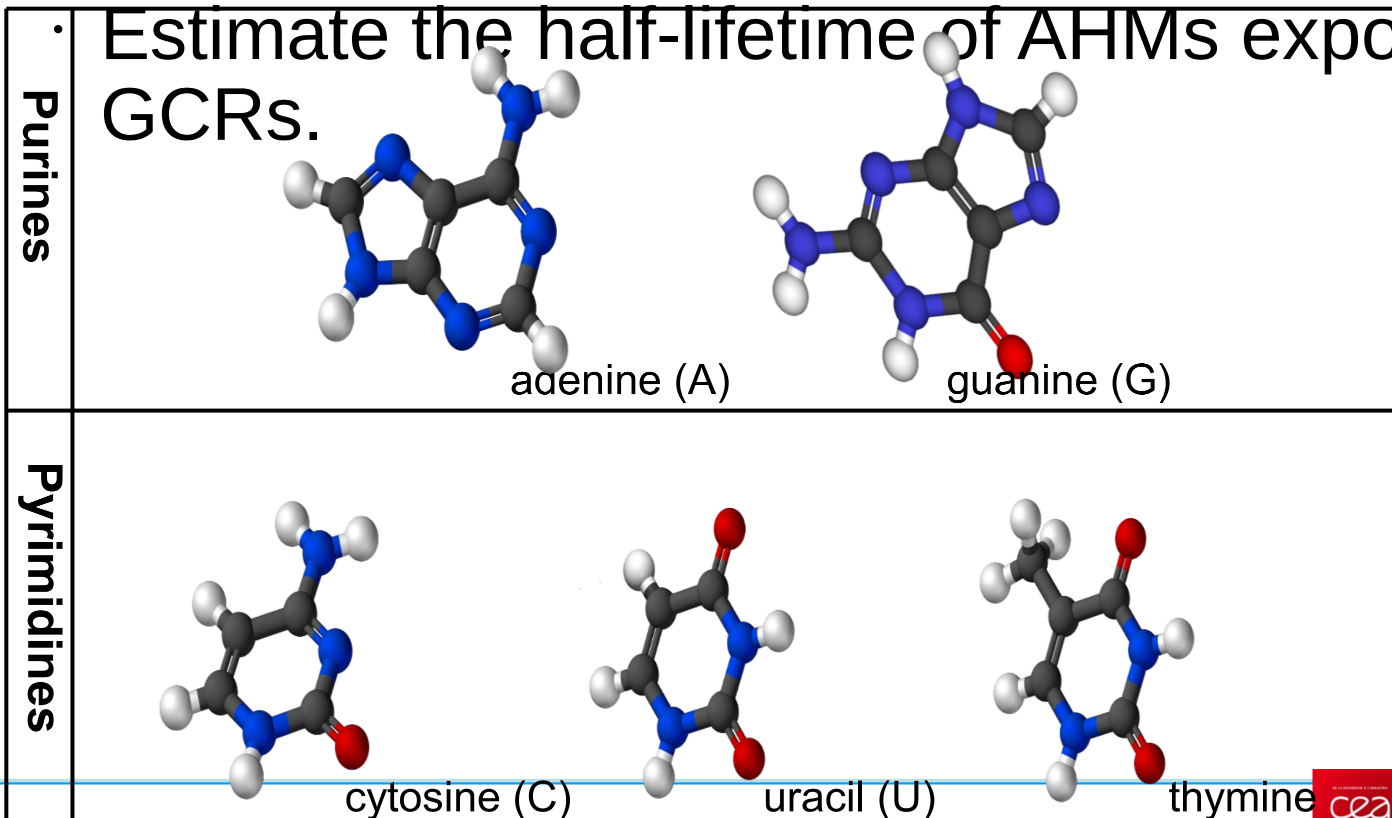
Aliphatic molecules are more radiosensitive than aromatic molecules.

Presence of AHMs in carbonaceous meteorites found on Earth.

https://nai.nasa.gov/annual-reports/2011/gsfc/research-activities-in-the-astrobiology-analytical-laboratory/

Objectives – What we do not know about the stability of AHMs under ionizing radiation.

- Study, at low temperature (~ 12 K), the effects of Galactic Cosmic Rays (GCRs) analogues (SHI) on solid samples of AHMs.
- Estimate the half-lifetime of AHMs exposed to GCRs.



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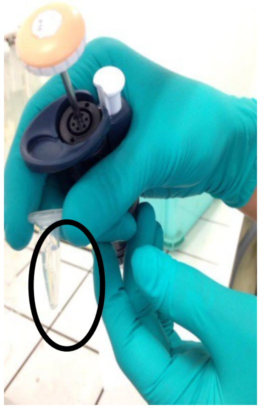
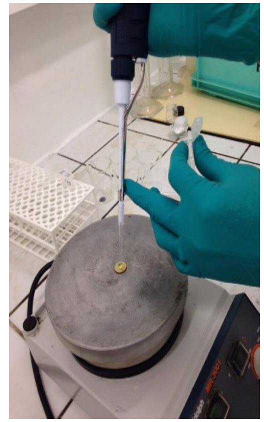
Sample preparation

Method 1



Nucleobase solution in water and ethanol



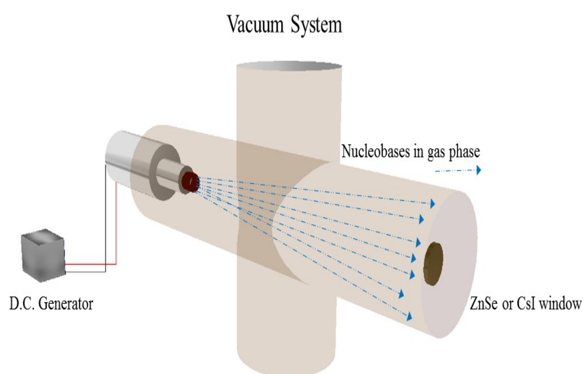
Drops of the solution onto a ZnSe window (heated to 100°C)

”Grainy” Sample

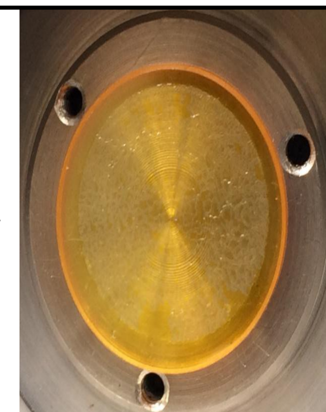
About 10 min

Method 2

Oven filled with nucleobase powder



Sublimation of the nucleobase powder by heating in high vacuum

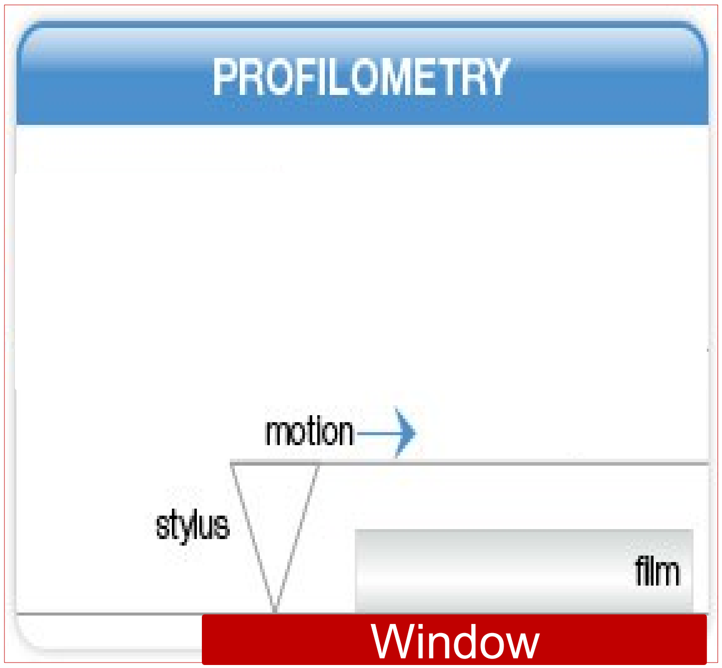


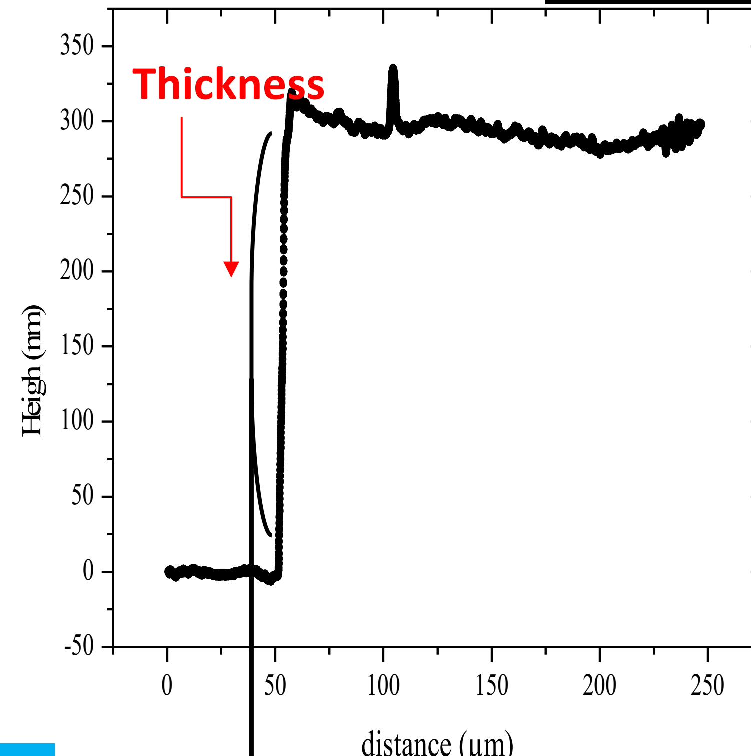
“Film” sample

Between 2 to 5 hours

Results – Profilometry experiments

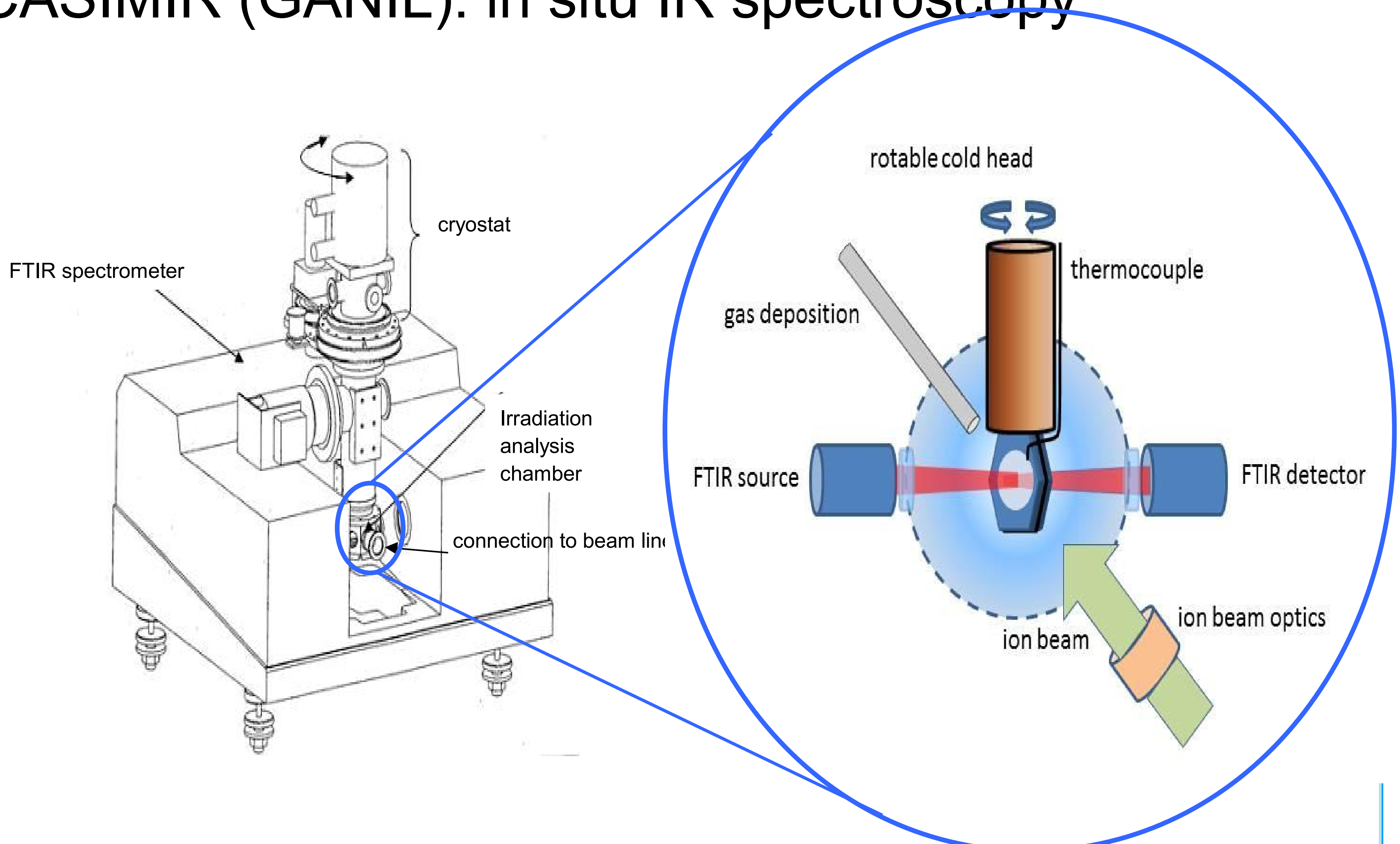
[redacted]





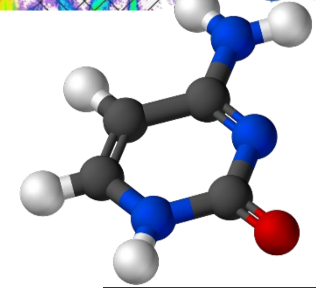
Thickness
Rugosity
Optical constant (absorption)

CASIMIR (GANIL): in situ IR spectroscopy



The set-up at GSI is similar

Results – Nucleobases radiolysis C4H5N3O

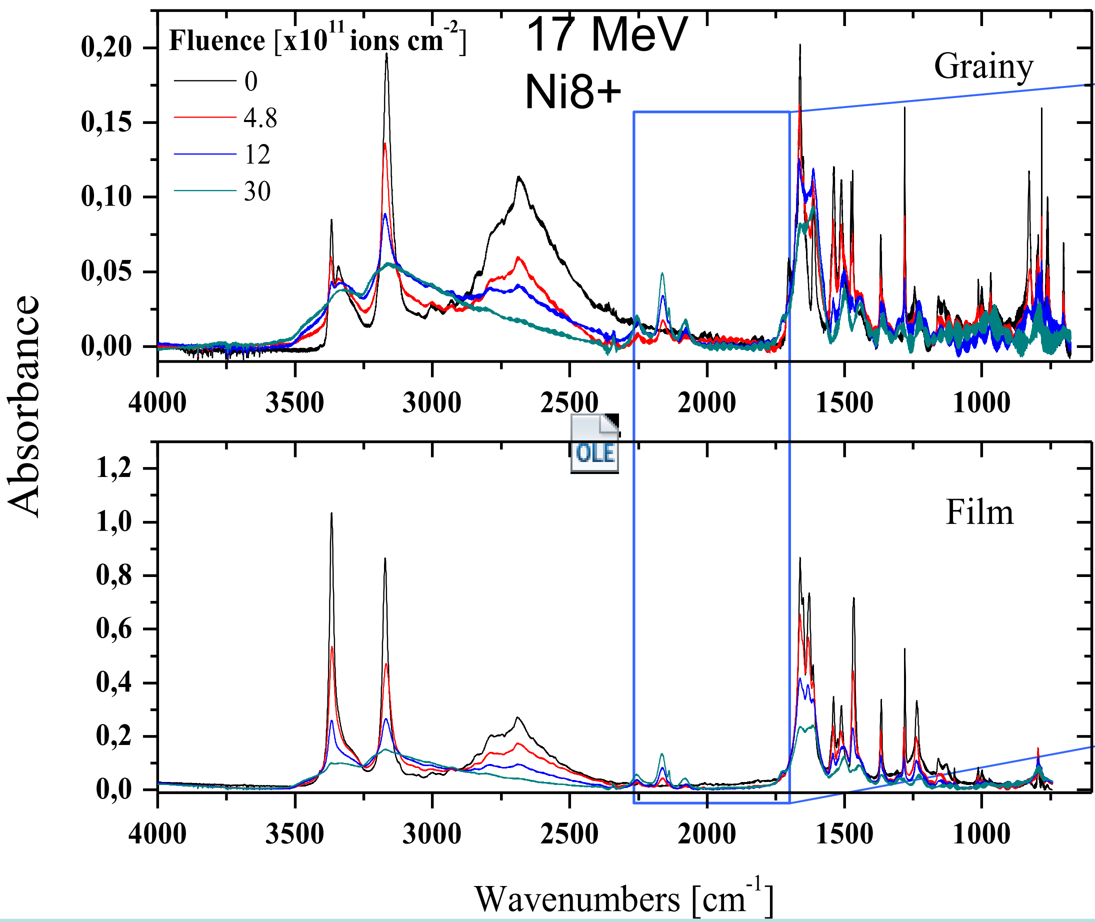


Summary of target and projectile characteristics

Ion Beam	Energy (MeV)	Electronic stopping power (keV.µm-1)	Nuclear stopping power (keV.µm-1)	Thickness (µm)	Penetration depth (µm)
U32+	116	1.34 x 104	2.5 x 102	0.34	17
Xe23+	92	1.13 x 104	7.3 x 101	0.8	6.5
Ni24+	632	4.80 x 103	4.4 x 101	0.28	137
Ni24+	632	4.80 x 103	4.4 x 101	0.20	137
Ca10+	190	3.10 x 103	2.3 x 100	1.1	9.1

nom

Evolution of samples under irradiation



[redacted]

“Grainy” and “film” samples under irradiation:

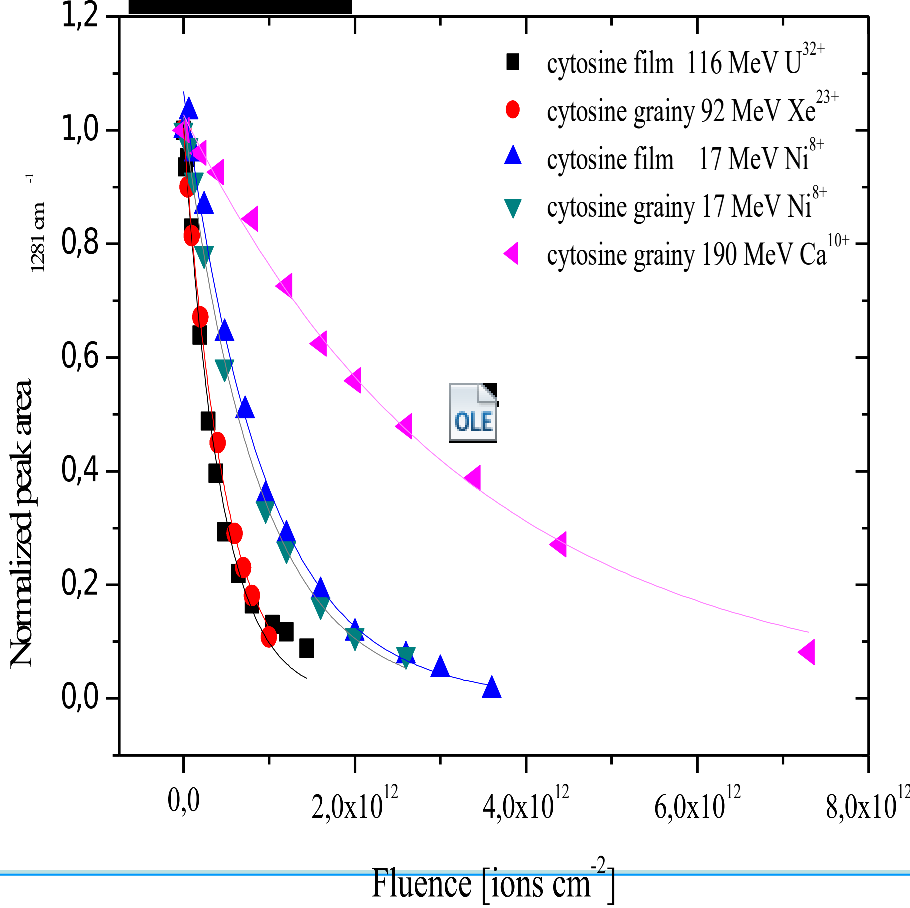
Decrease of the peak area intensity

New absorption peaks in the same region

nom

Evolution of samples under irradiation

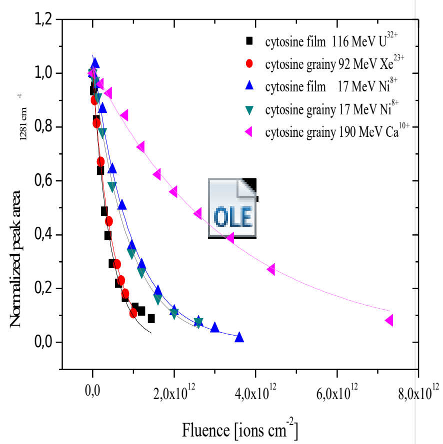
$$A = A_0 e^{-\sigma_d F}$$

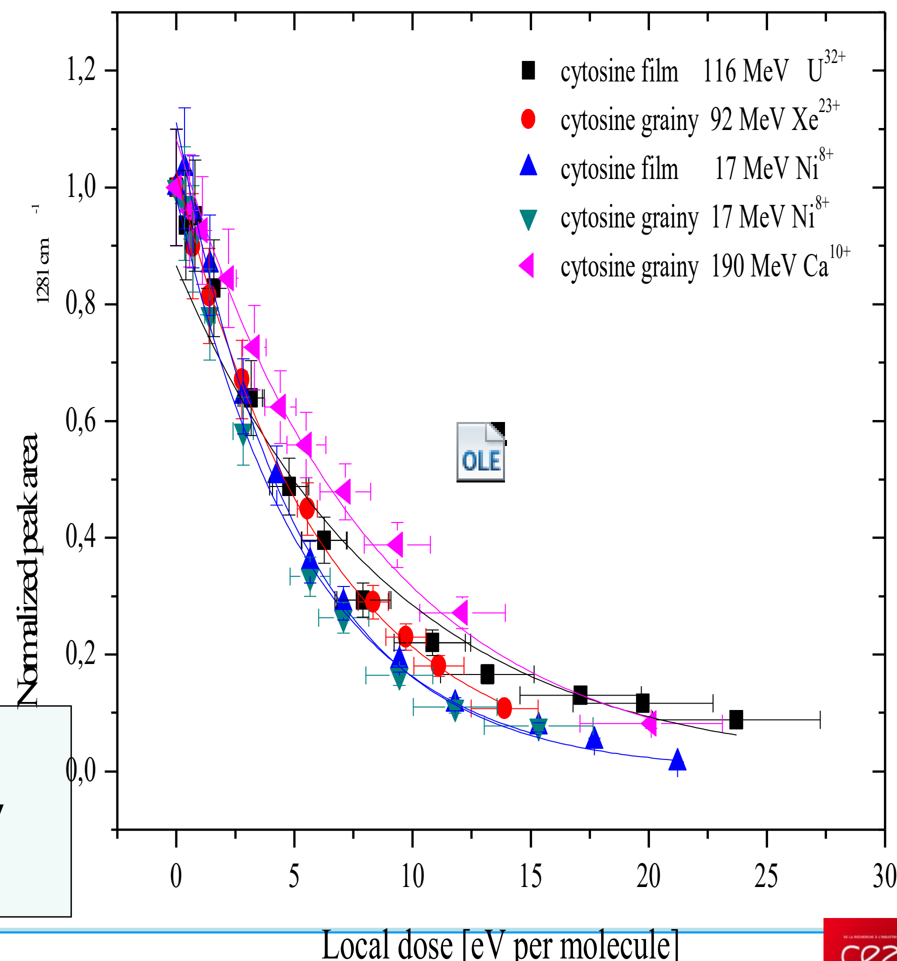


Sample	Destruction cross section (x10-13 cm2)	Se [x103 keV µm-1]	
Ca10+	0.57 ± 0.01	3.2	◀
Ni8+	1.31 ± 0.06	4.8	▼
Ni8+	1.17 ± 0.03	4.8	▲
Xe23+	3.20 ± 0.3	11.3	● ■
U32+	3.59 ± 0.09	13.4	

Evolution of samples under irradiation

[redacted]





Key parameter
LOCAL DOSE (energy per molecule)

nom

All nucleobases were exposed to 190 MeV Ca10+ at GSI

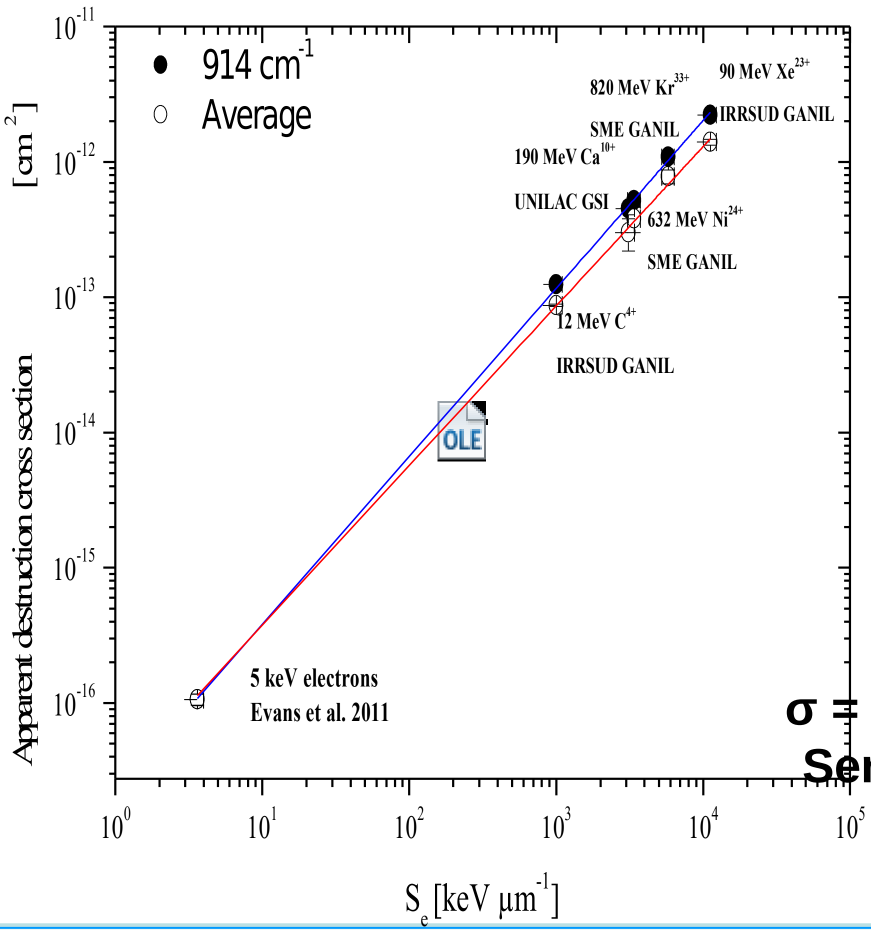
$$\sigma_{guanine} < \sigma_{adenine} < \sigma_{thymine} \approx \sigma_{uracil} \approx \sigma_{cytosine}$$

$$\sigma_{Purine} < \sigma_{Pyrimidine}$$

Purine nucleobases are more radioresistant than the pyrimidine nucleobases

AHM	Average destruction cross sections [× 10-13 cm2]	Projectile
Guanine	(1.3 ± 0.8)	190 MeV Ca10+
Adenine	(3.0 ± 0.8)	190 MeV Ca10+
Thymine grainy	(4.0 ± 1)	190 MeV Ca10+
Thymine film	(5.5 ± 0.8)	190 MeV Ca10+
Cytosine	(5.0 ± 1)	190 MeV Ca10+

Apparent destruction cross section as a function of the electronic stopping power [redacted]



σ = C Sen

$$\sigma = (2.0 \pm 0.4) \times 10\text{-}17$$
$$Se(1.24 \pm 0.01)$$

Experiments performed in two different laboratories fall on the same curve. (GANIL and GSI).

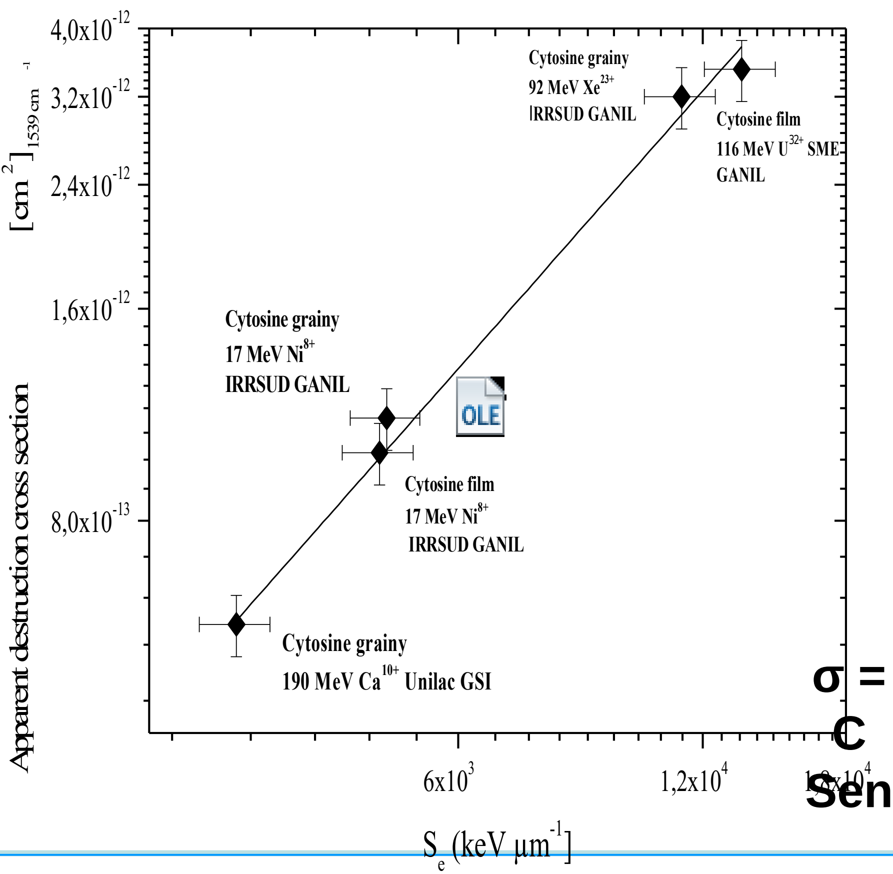
Vignoli Muniz et al., Astrobiology 17, 2017

nom

Apparent destruction cross section as a function of the electronic stopping power

[redacted]

$$\sigma_d = (1.5 \pm 1.0) \times 10^{-17} S_e^{(1.3 \pm 0.08)}$$



σ = C Sen

All results obtained by our group:

$$1 \leq n \leq 1.5$$

experiments performed in two different laboratories fall on the same curve.
(GANIL and GSI).

nom

Astrophysical implications

Estimation of nucleobase survival time

Half-life of solid adenine exposed to cosmic rays in the ISM.

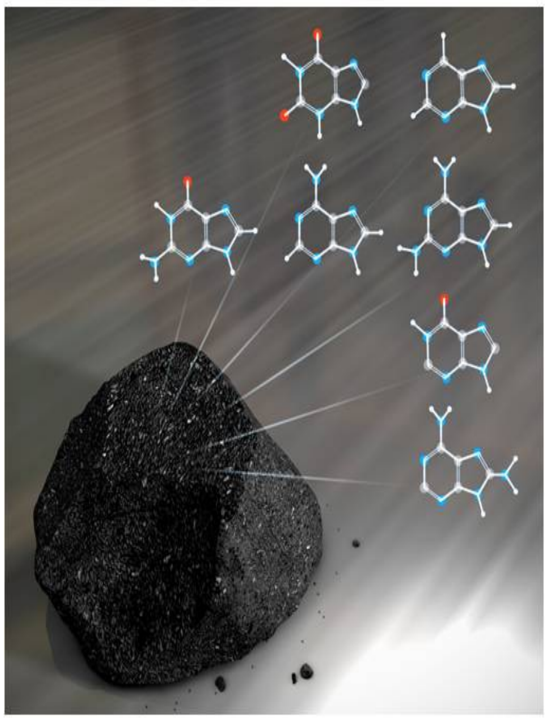
$$\tau_{1/2} = \ln 2 \left(4\pi \sum_Z \int_{10^{-1}}^{10^{3}} \sigma(Z,E)\, \Phi(Z,E)\, dE\right)^{-1}$$

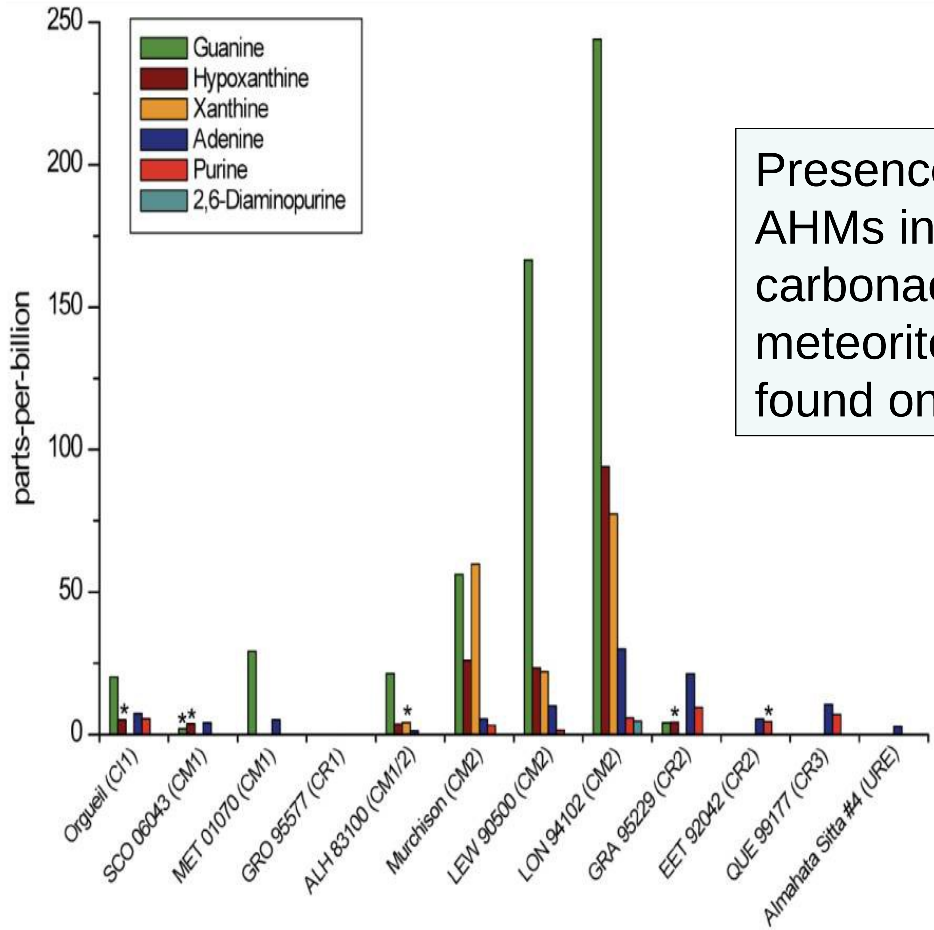
$$\tau_{1/2} = (14 \pm 11)\ \text{Myears}$$

Dense Clouds: average time of survival = 10 Myears

High survival probability!

Relatively high presence of guanine in carbonaceous meteorites





Presence of AHMs in carbonaceous meteorites found on Earth.

nom

https://nai.nasa.gov/annual-reports/2011/gsfc/research-activities-in-the-astrobiology-analytical-laboratory/

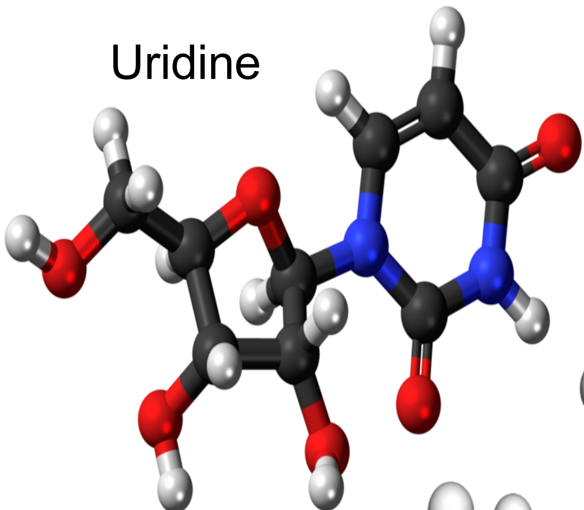
Outlooks:

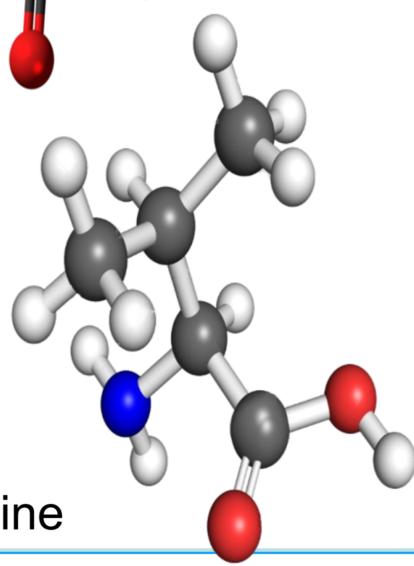
- Irradiation of simple ices : CH_3OH,…
- Interfaces : C/Ices Silicates/ices
- Different experimental approaches are necessary to monitor the evolution of ices and complex molecules under SHI irradiation:
  - TOF – SIMS (mass spectrometry, secondary ion emission)
  - QCM (Quartz-Microbalance, total sputtering)
- Study AHMs in more realistic conditions, i.e., in water matrix.
- Since PAHs are an important source of carbon in outer space, radioresistance/ reactivity after irradiation.
- Study the stability of other complex molecules

Uridine and d-valine were irradiated at room temperature.

The data is under analysis at the time of this pr

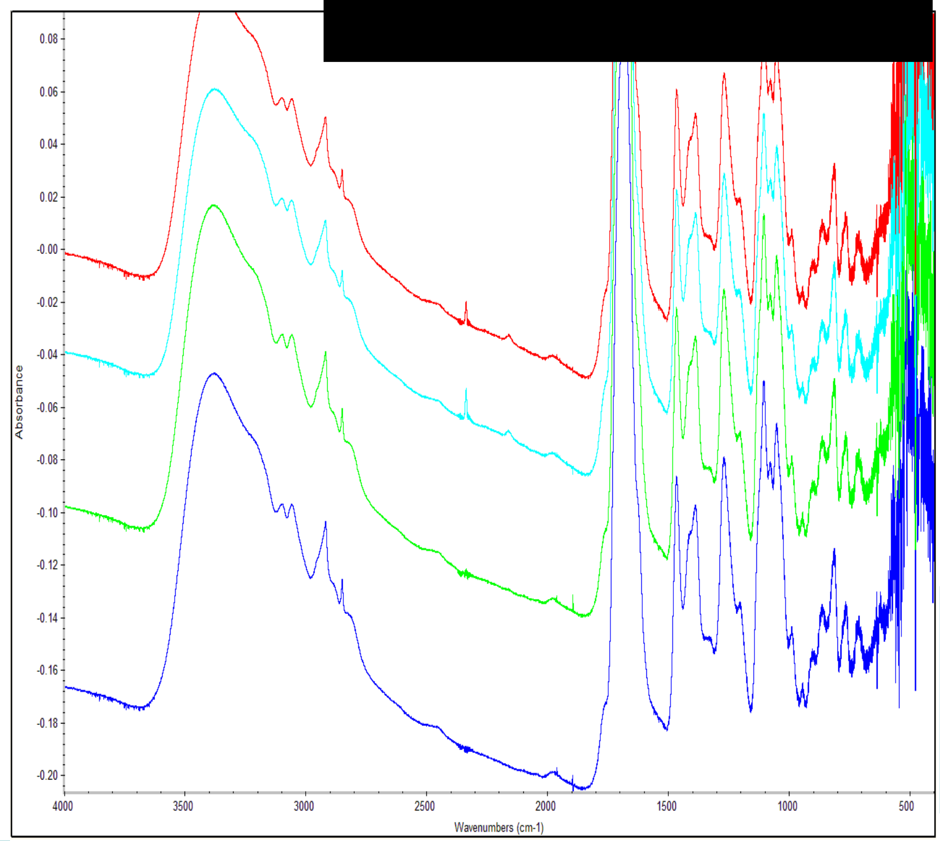
Uridine





D-Valine

[redacted]



nom

Thank you

Jingjie, Xueyang, Gianni, Thomas, Hermann, Philippe, Stéphane

Vinicius, Ana, Sergio

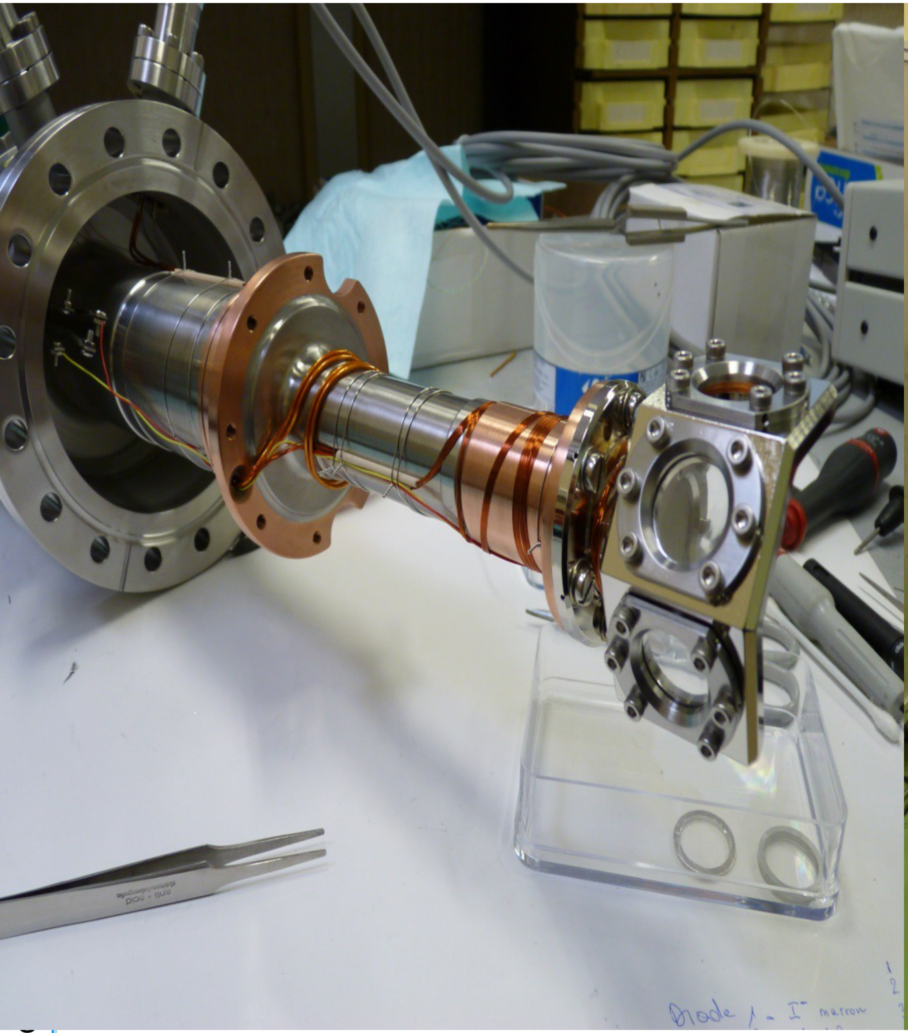
Hussein+Enio

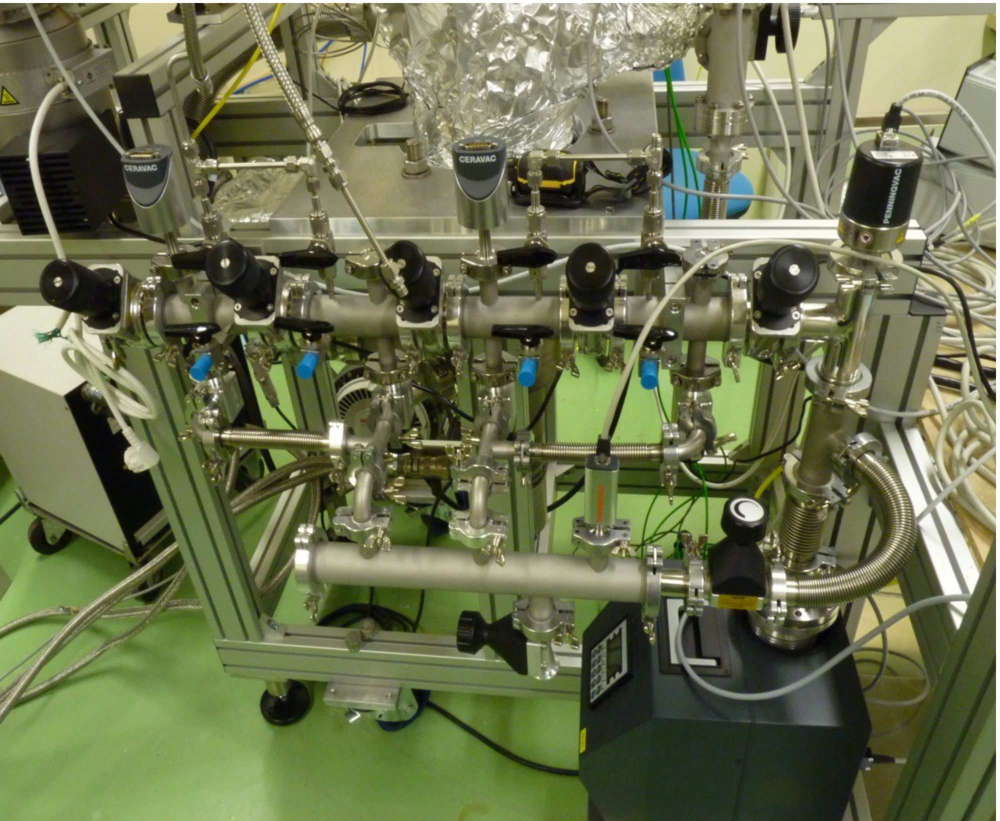
Eduardo

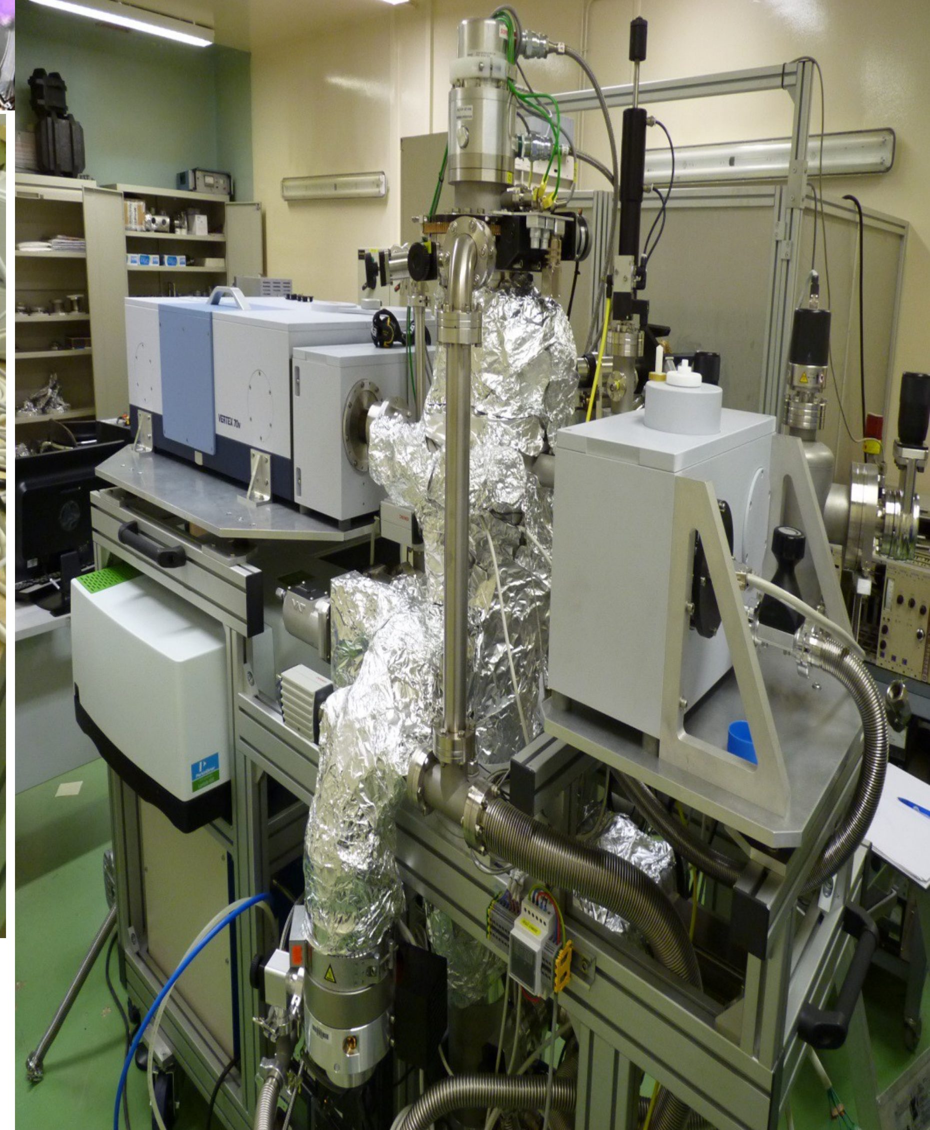
Emmanuel

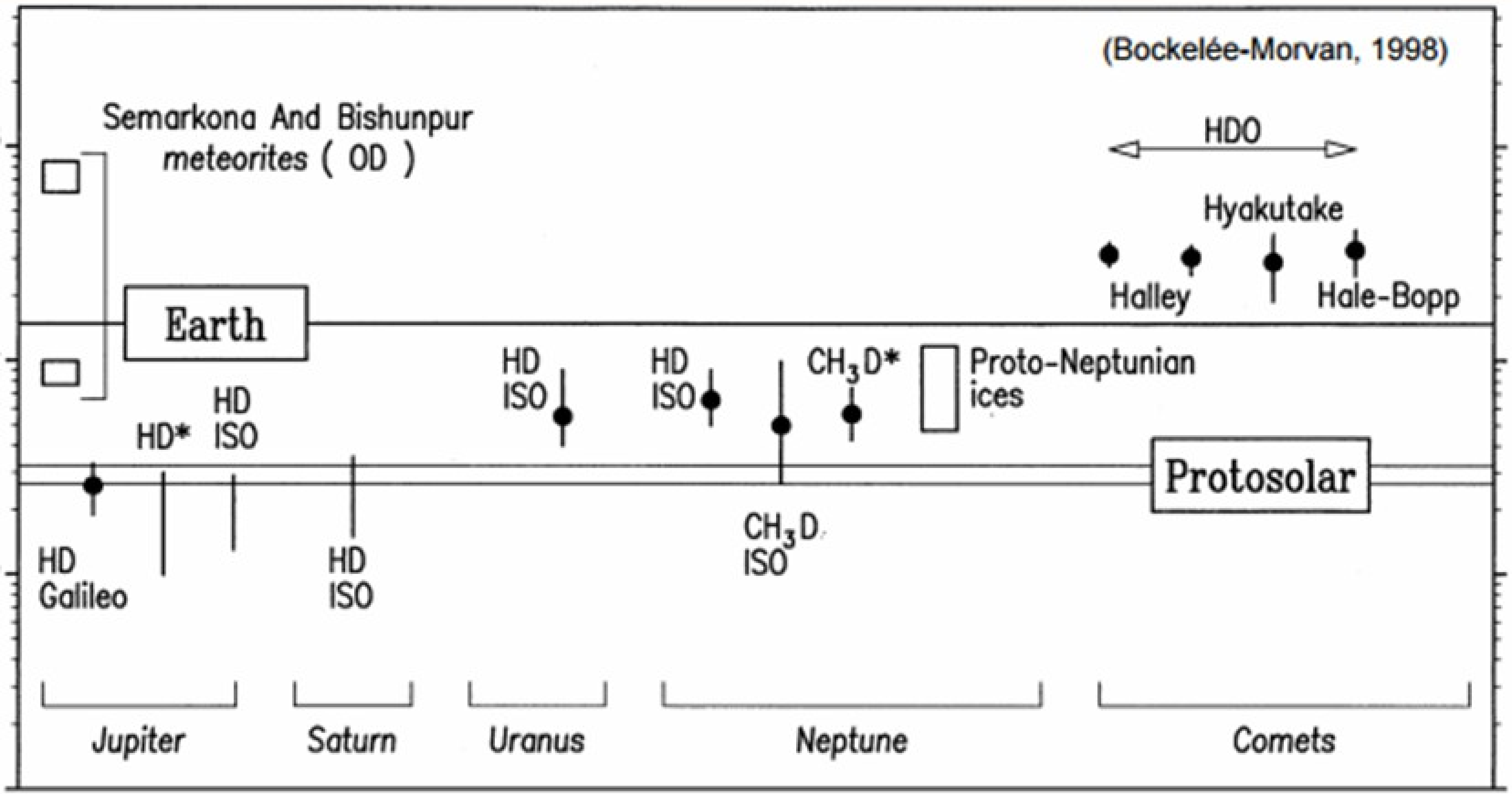
Alicja

Some pictures:

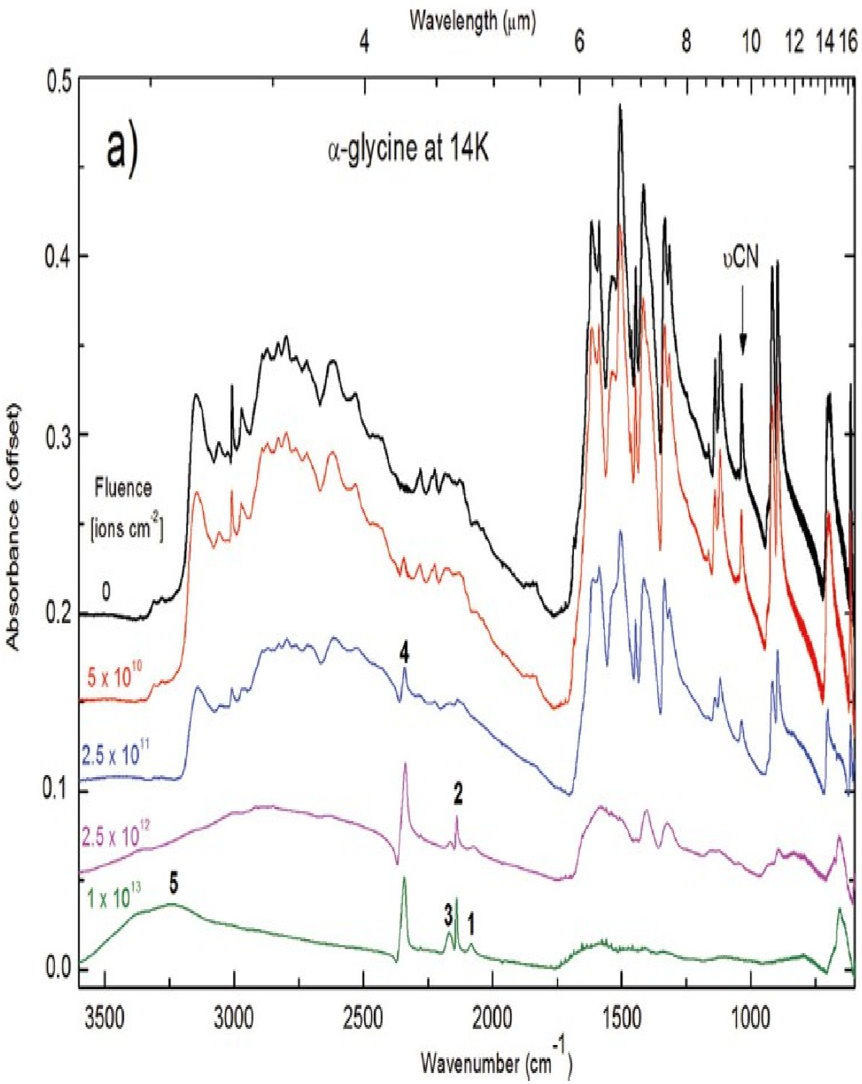


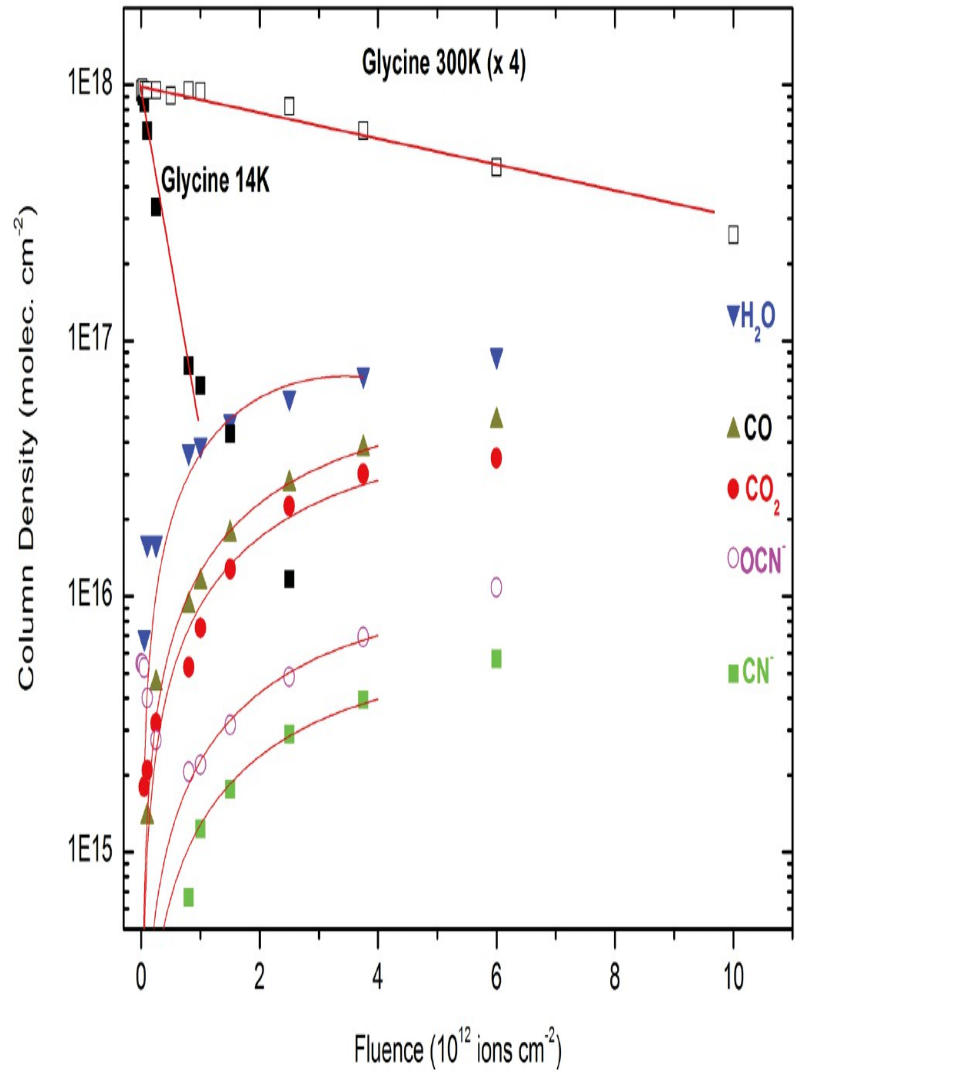

CERAVAC
CERAVAC
PENNINGVAC




(Bockelée-Morvan, 1998)
Semarkona And Bishunpur
meteorites (OD)
D / H
10^{-3}
10^{-4}
10^{-5}
Earth
Protosolar
HDO
Hyakutake
Halley
Hale-Bopp
HD
Galileo
HD*
HD
ISO
HD
ISO
HD
ISO
HD
ISO
CH_3D*
CH_3D
ISO
Proto-Neptunian
ices
Jupiter
Saturn
Uranus
Neptune
Comets

Glycine at 14K and 300 K 58Ni11+@ 46MeV



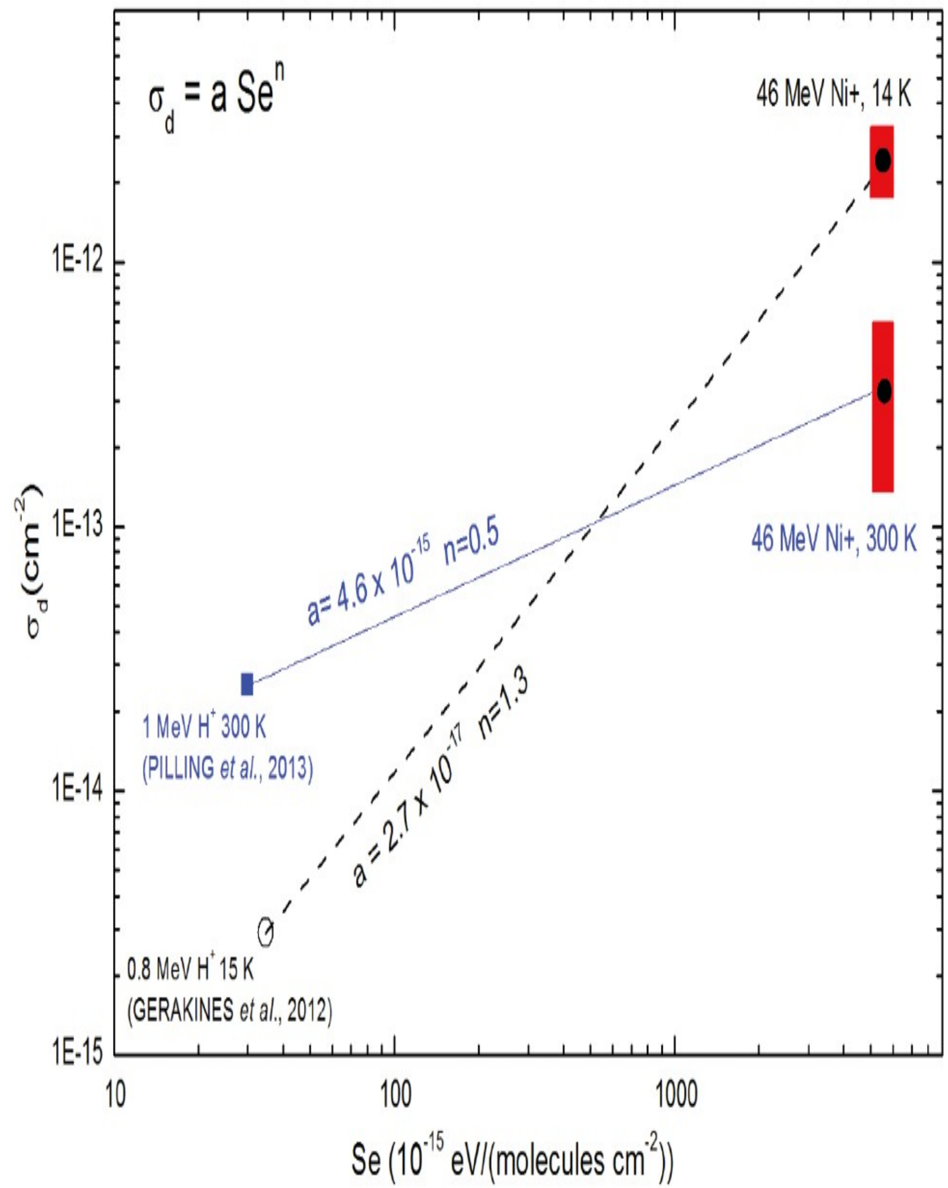


Destruction cross section:

σ=2,4 10-12 cm2 @ 14K

σ=3,4 10-13 cm2 @ 300K

Temperature effect



σ=ASen

Temperature (K)	n
14	1.3
300	0.5

Centre de Recherche sur les Ions, les Matériaux et la Photonique