How (heavy ion) **radiation affects interstellar and solar system ice mantles**

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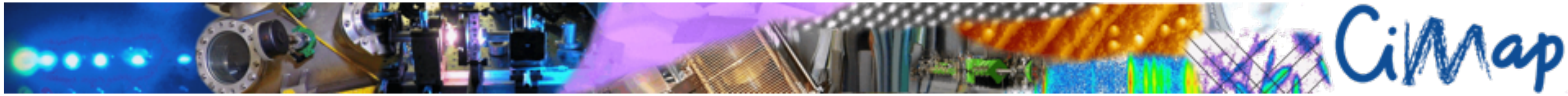


astrophysical materials:

carbonaceous, silicates, and **Ices**

laboratory simulation: **A**

solar wind, ions trapped
in magnetospheres,
and ***cosmic rays***

at heavy **ion accelerators**
GANIL Aribe, Irrsud, SME, HE
+ GSI Unilac: from **keV** to **GeV**

Infrared absorption spectroscopy FTIR

1st step: ***physics:***
Compaction, amorphization, **sputtering**
radiolysis: fragmentation (destruction)

2nd step: ***chemistry:***
formation of new molecules
(radicals, "Implantation")

astrophysics:

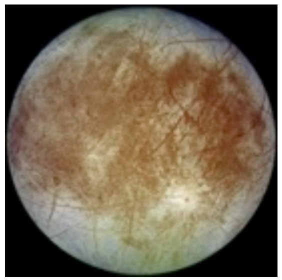
Amorphization + Compaction
of Water Ice H_2O **B**

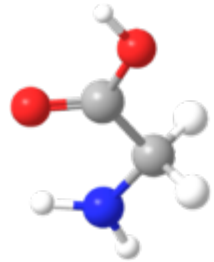
Radiolysis of CO + CO_2
Origin of gas phase molecules
in interstellar dense clouds ? **C**
... cosmic rays versus UV photons

astrochemistry:

Origin of CO_2 and H_2SO_4 on Europa?
endogenic, or exogenic
= implantation of C and S ions ? **D**

Formation and radiation resistance
of organic/prebiotic molecules **E**

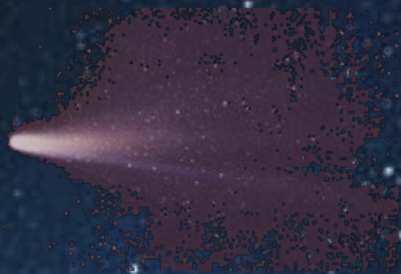




Astrophysical Ices ...

... exposed to irradiation by UV photons, electrons, and ions:
Cosmic rays, Solar Wind, Giant Planet's Magnetospheres

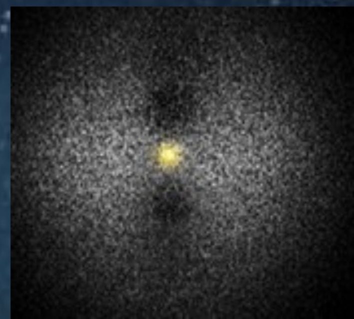
→ Comets



→ Giant Planet's Moons
(Europa, Ganymede, ...)

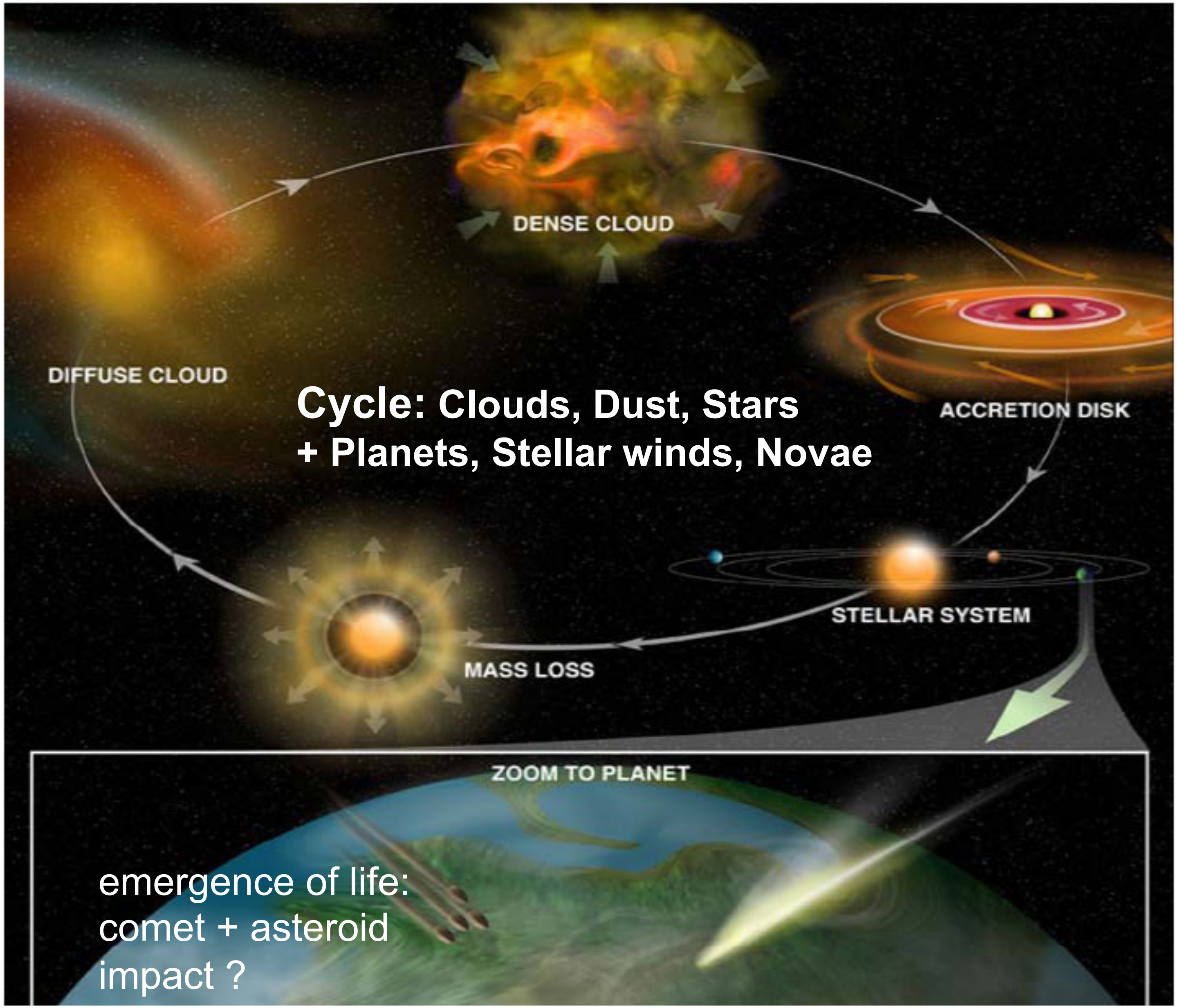

→ Dust Grains

Rings




Dense Interstellar Clouds
(birthplaces of suns and planets)

Astrochemistry and Astrobiology



Interstellar Medium: Dense Molecular Clouds

Density of 10^3-10^6 particles cm^{-3}, mainly H_2, T~10 K. Star formation.

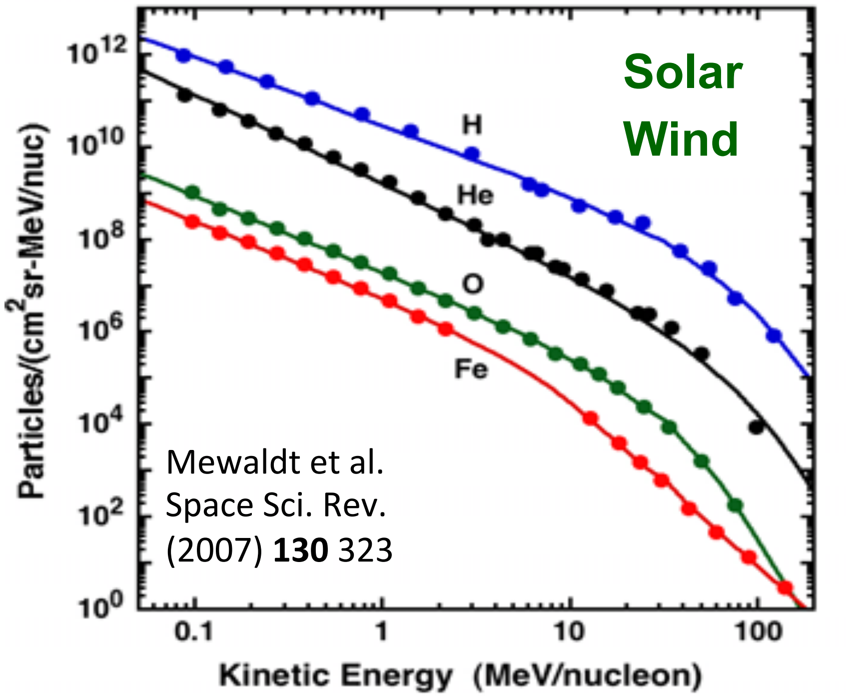
Molecules in gas phase and dust grains covered by ice mantles.

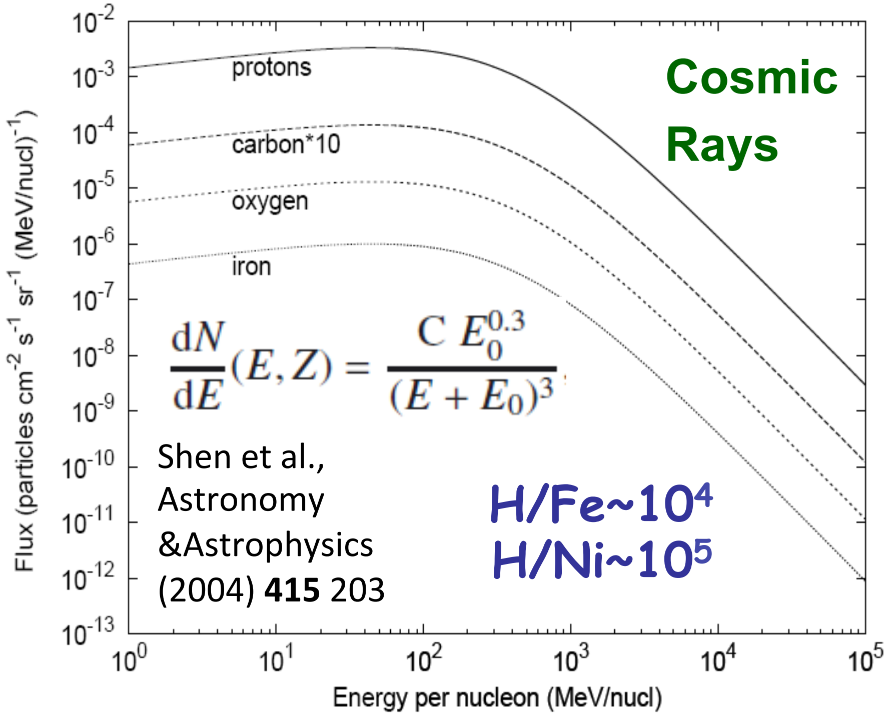
Possibly complex organic chemistry in ice mantles due to:
- Surface reactions
- UV and ion processing.

Horsehead Nebula

Size: up to some tens of Parsecs
Lifetime: 10^6-10^9 years
freeze-out time: $10^9/n_H$ (years)

Radiation Field in Space : complex ! (UV, e-, x-rays, ions)



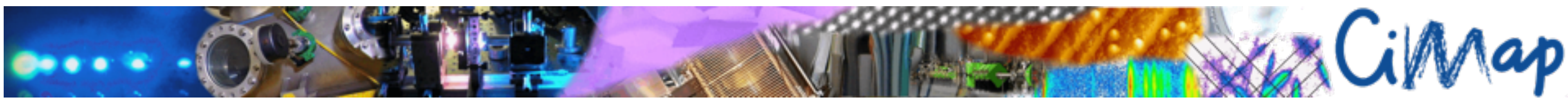


Heavy Ions: why? - large electronic energy loss S_e

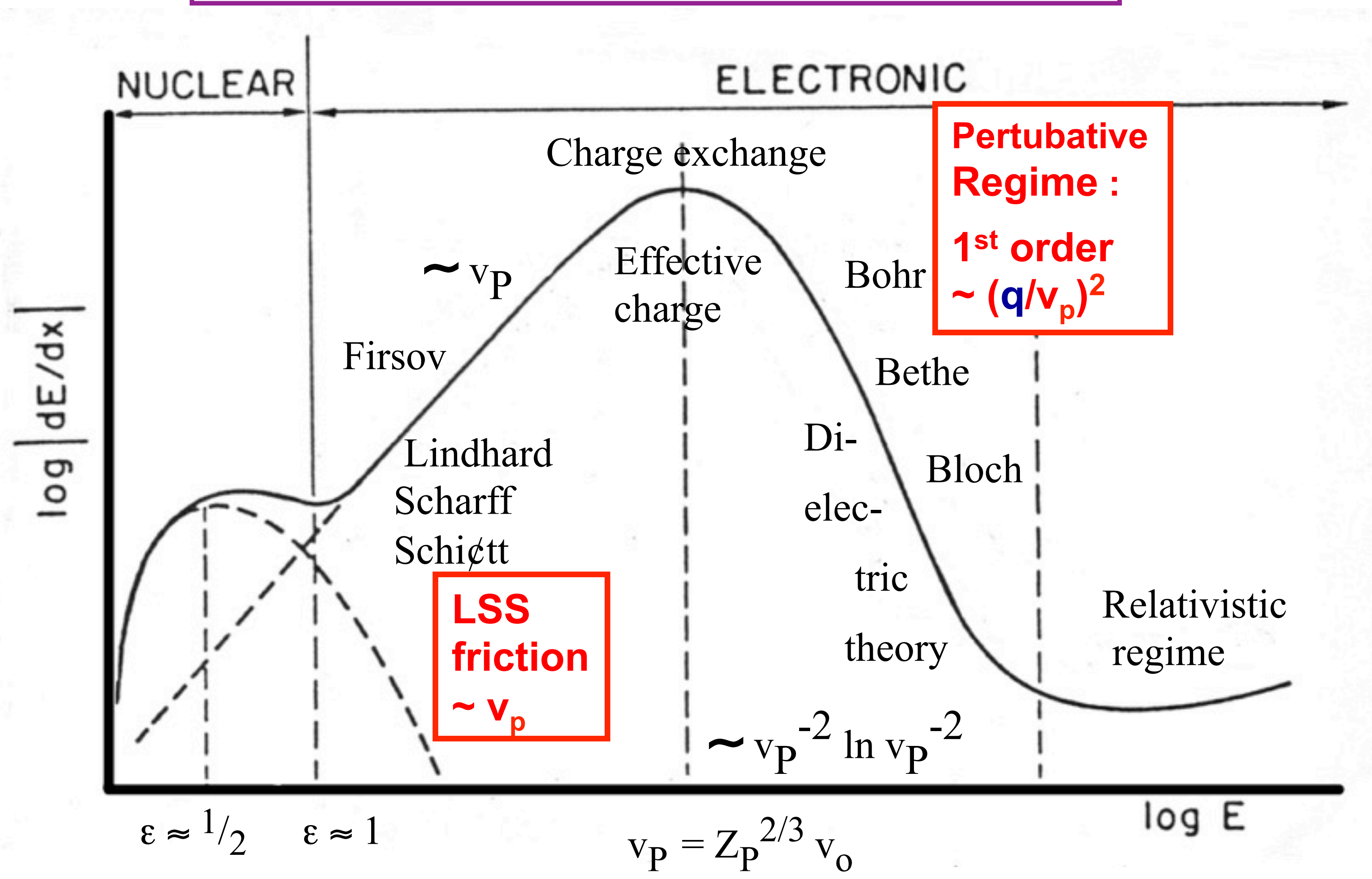
- Scaling laws: S_e^n with n ≈ ½,1,$^3/_2$,**2**, ... 4)

- Unexplained findings (gas phase molecules in dense clouds...), few data

- Astrochemistry: origin of CO_2 and H_2SO_4 on Europe, emergence of life?



Energy loss as a function of projectile energy

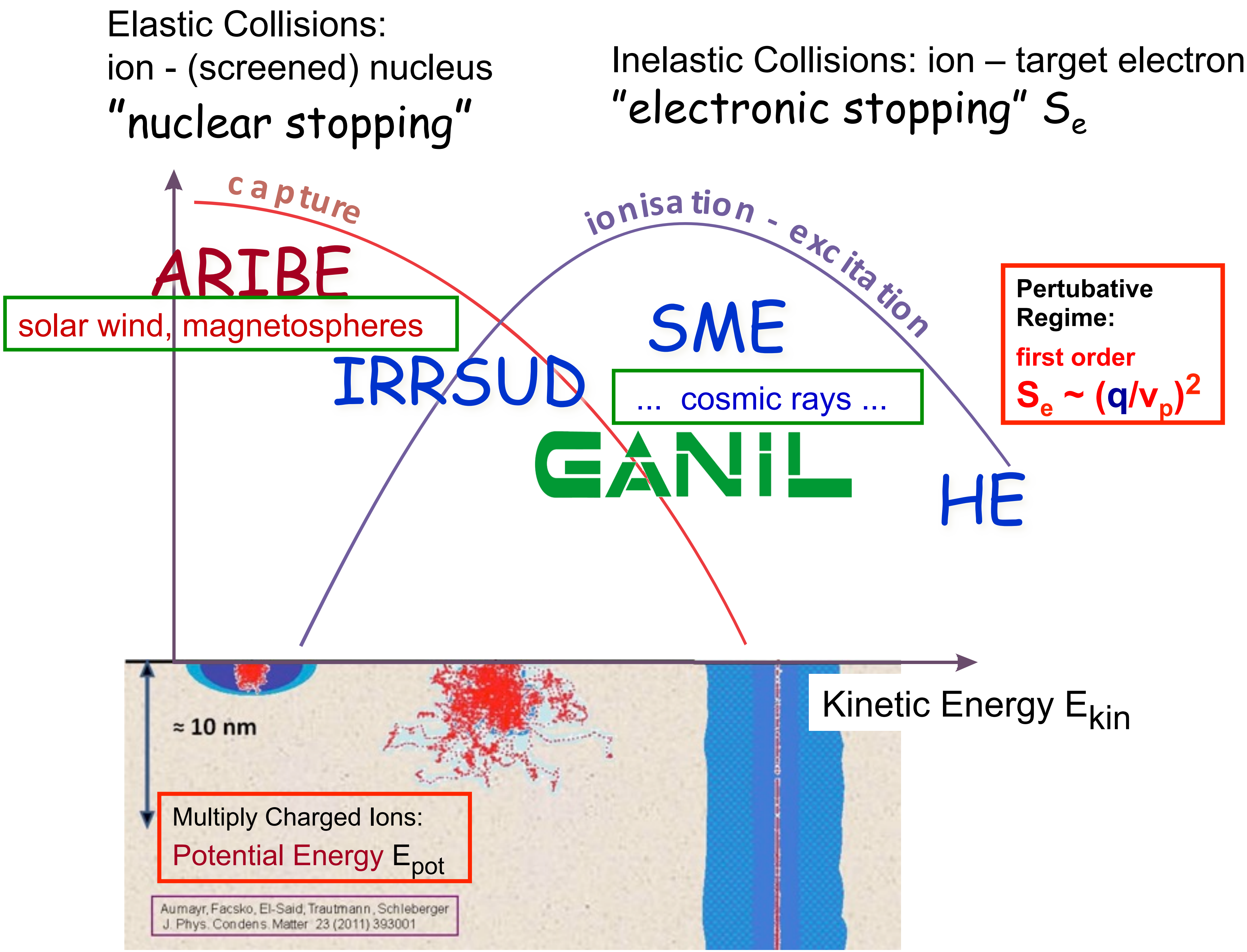


solar/stellar wind

cosmic rays

A

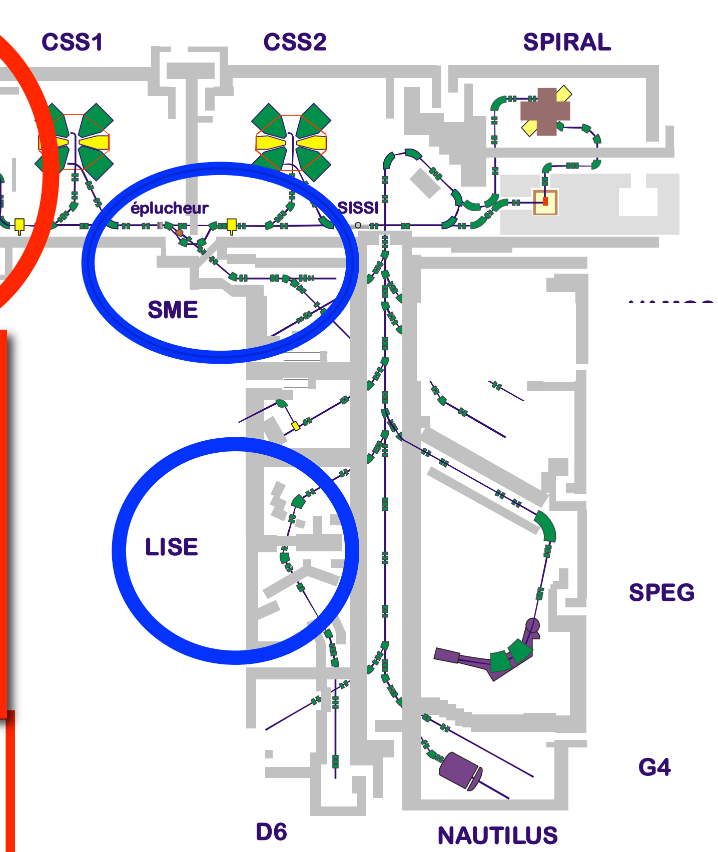
Astrophysics and chemistry in the laboratory: simulation of solar wind, cosmic ray and magnetosphere ion irradiation of astrophysical ices



Elastic Collisions:
ion - (screened) nucleus
"nuclear stopping"
Inelastic Collisions: ion – target electron
"electronic stopping" S_e
capture
ionisation - excitation
ARIBE
solar wind, magnetospheres
IRRSUD
SME
... cosmic rays ...
GANIL
HE
Pertubative Regime:
first order
$S_e \sim (q/v_p)^2$
Kinetic Energy E_{kin}
≈ 10 nm
Multiply Charged Ions:
Potential Energy E_{pot}
Aumayr, Facsko, El-Said, Trautmann, Schleberger
J. Phys. Condens. Matter 23 (2011) 393001

Astromaterials @GANIL

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 MeV/u

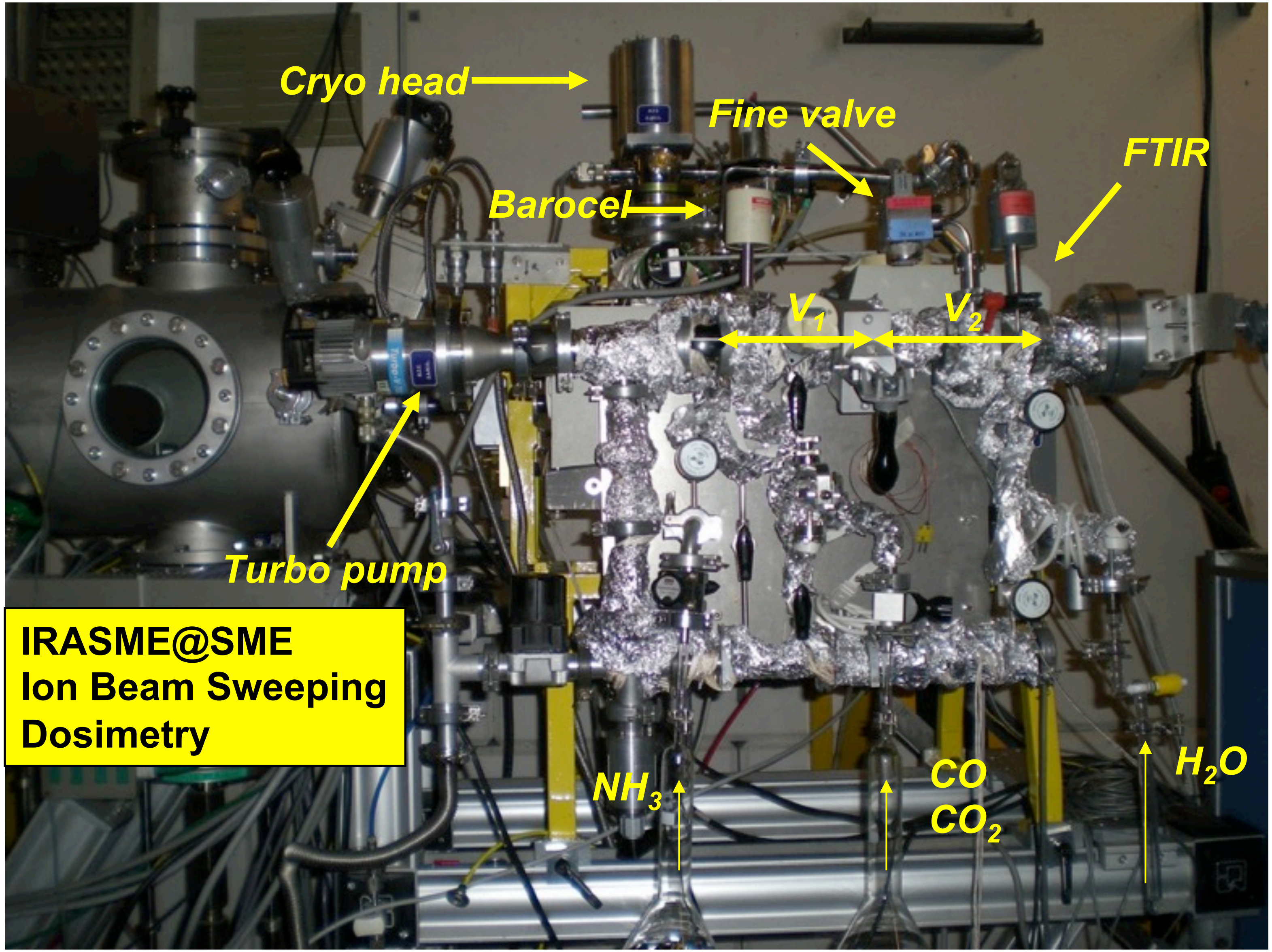
High Energy: LISE

Fe: 70 MeV/u

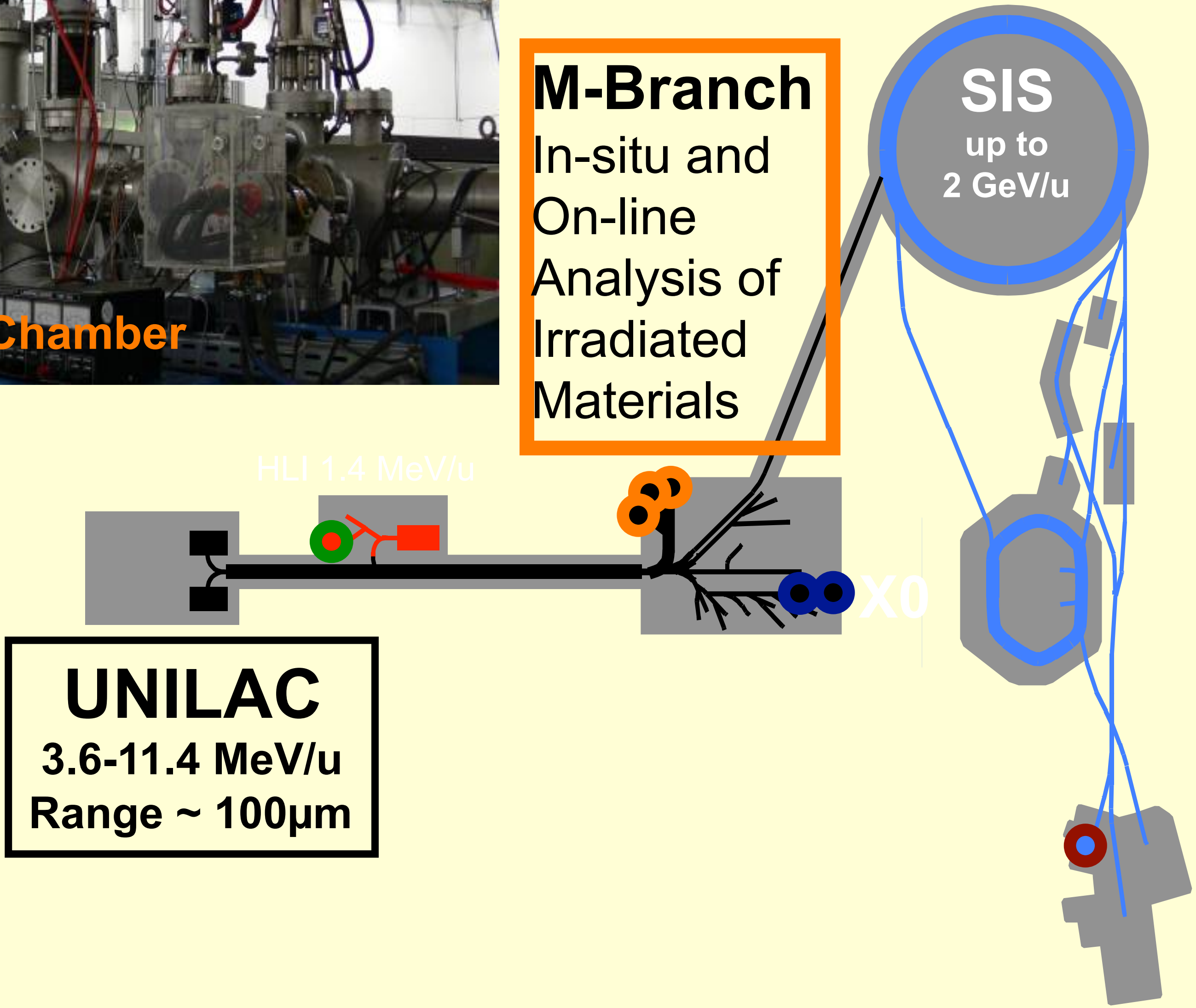
Medium Energy: SME

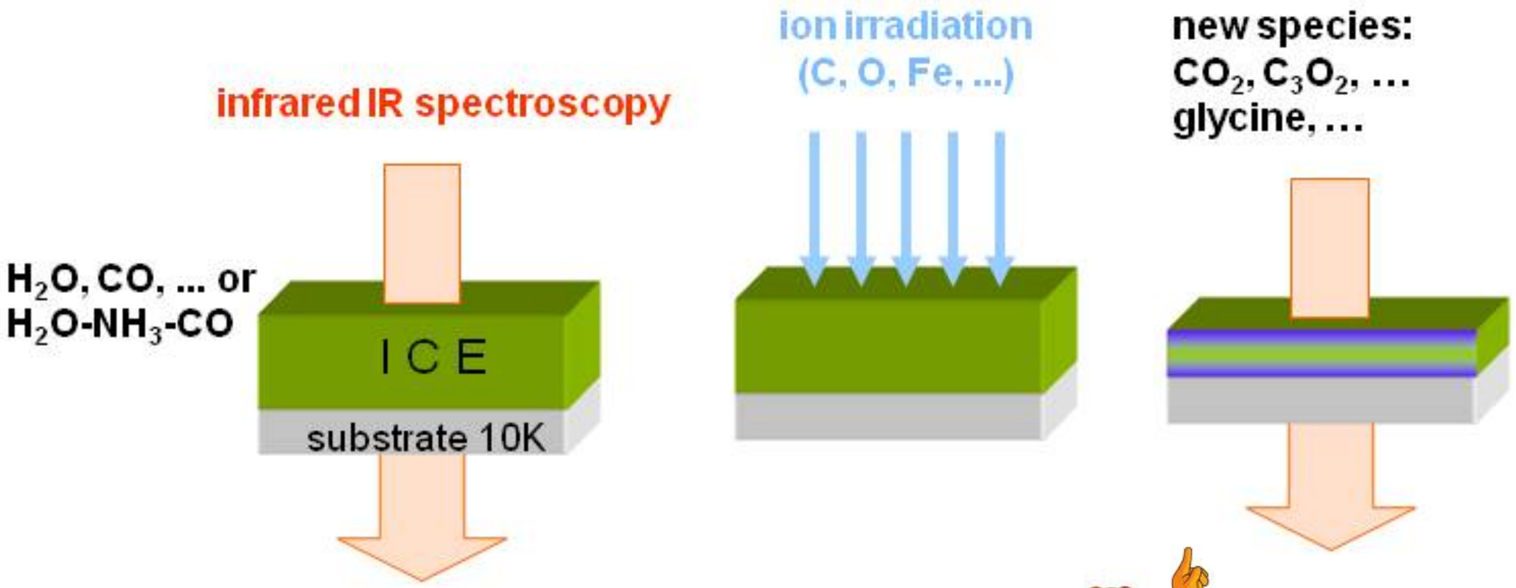
O, Fe, Ni, Kr: 5-13 MeV/u

the "gas mixing and deposition machine"



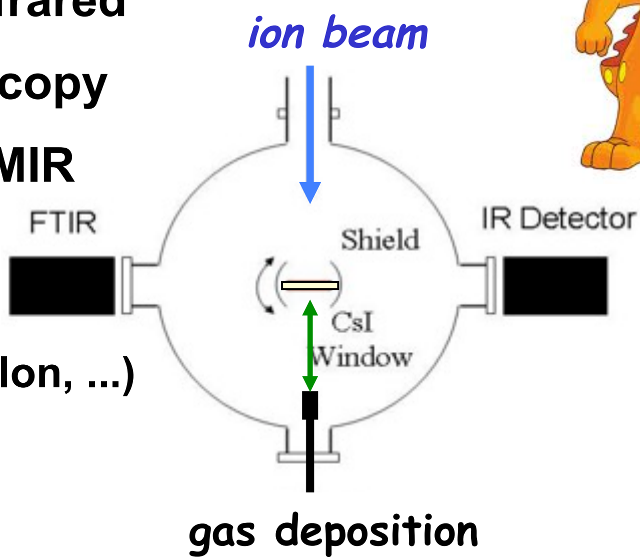
Materialforschung

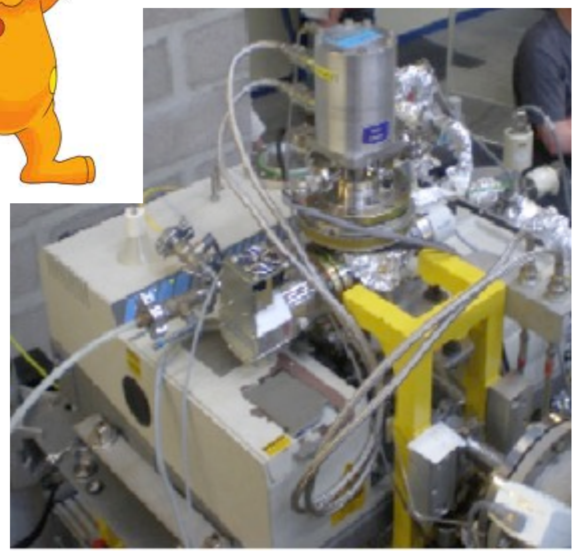




Fourier Transform Infrared Absorption Spectroscopy

FTIR @CIMAP: CASIMIR





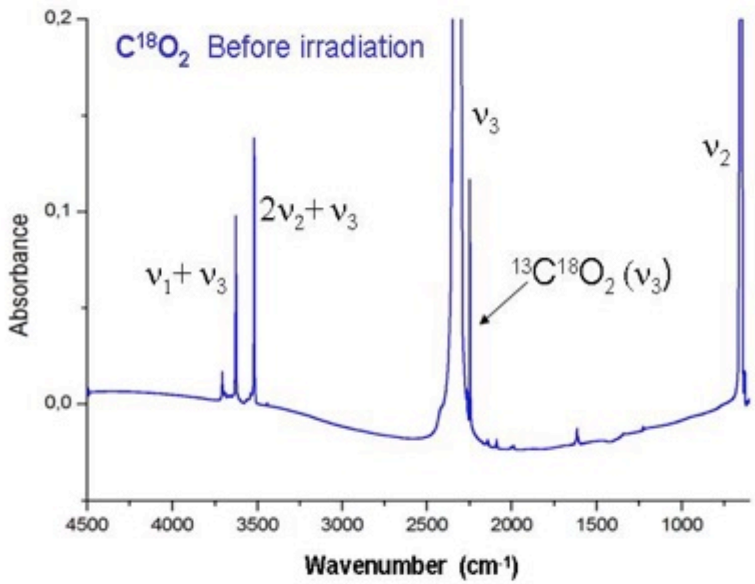
(E. Balanzat, J.M. Ramillon, ...)

FTIR Fourier Transform Infrared Absorption Spectroscopy: molecular vibrations

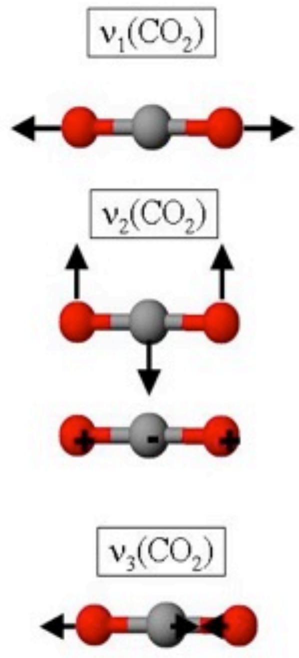
plenty of information:

Absorption Line Position + Shape:
identification of molecules, environment ("dangling bonds": porosity ...)
structure (crystalline, amorphous)

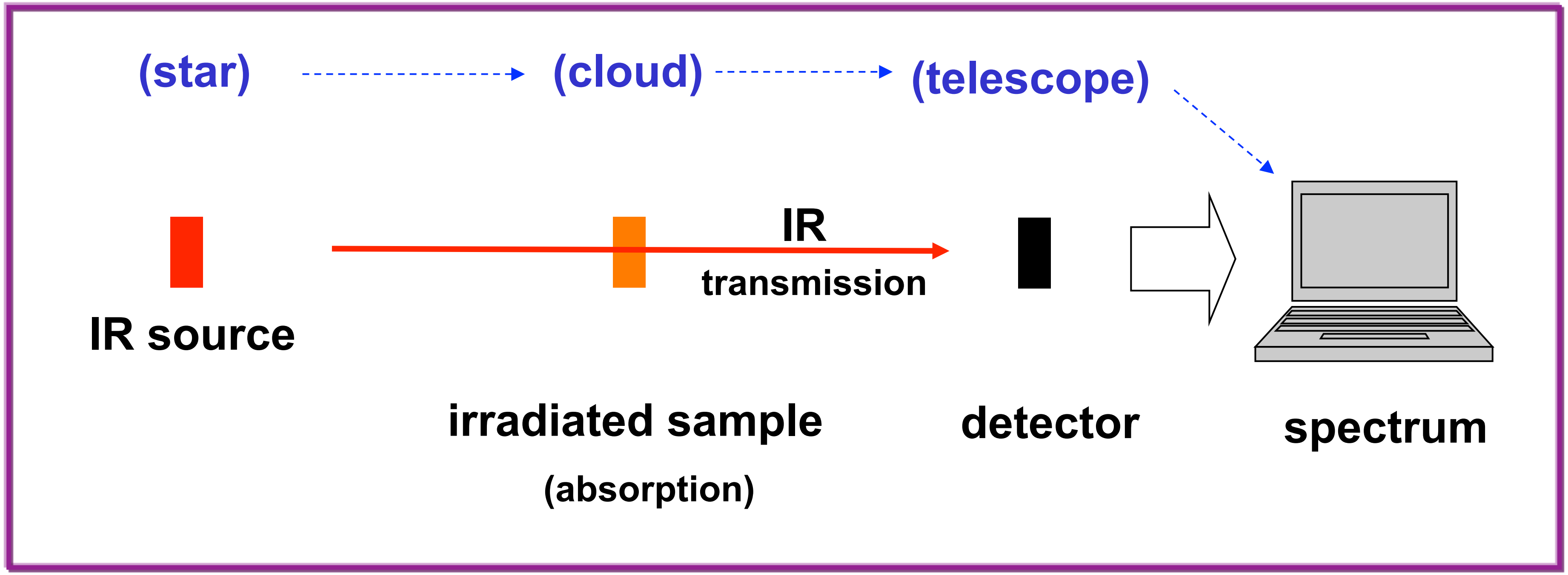
Integral (Surface)
columnar density (thickness)
evolution with projectile fluence: disappearance and synthesis of molecules

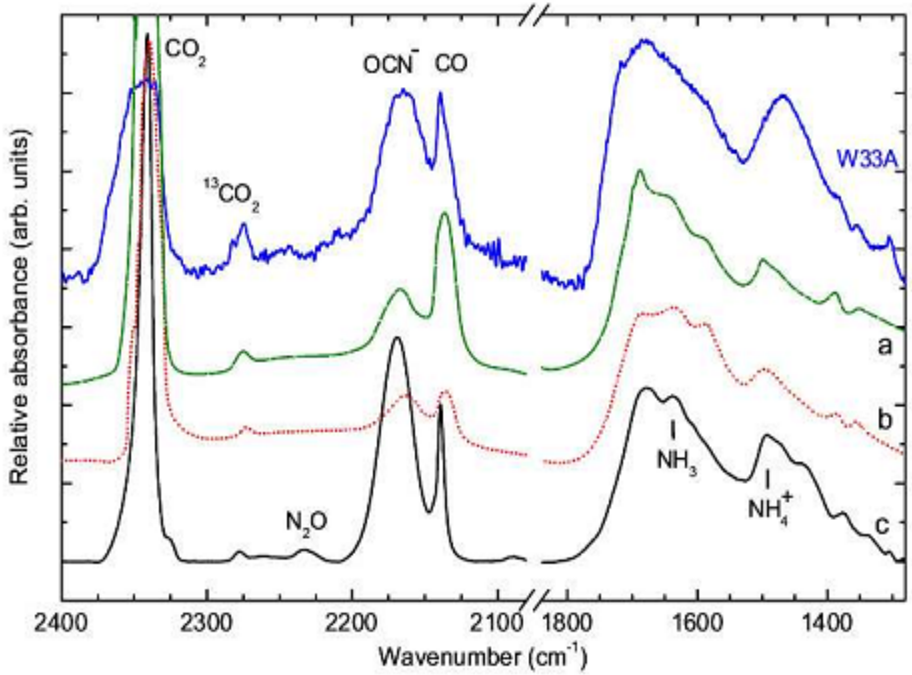


FTIR spectrum of $C^{18}O_2$ ice at 15 K



but:
detection of symmetric molecules (O_2, N_2 ...) difficult





Space observation:

ISO Infrared Space Observatory, protostellar source W33a

Laboratory simulation:

UV photons

protons

heavy ions

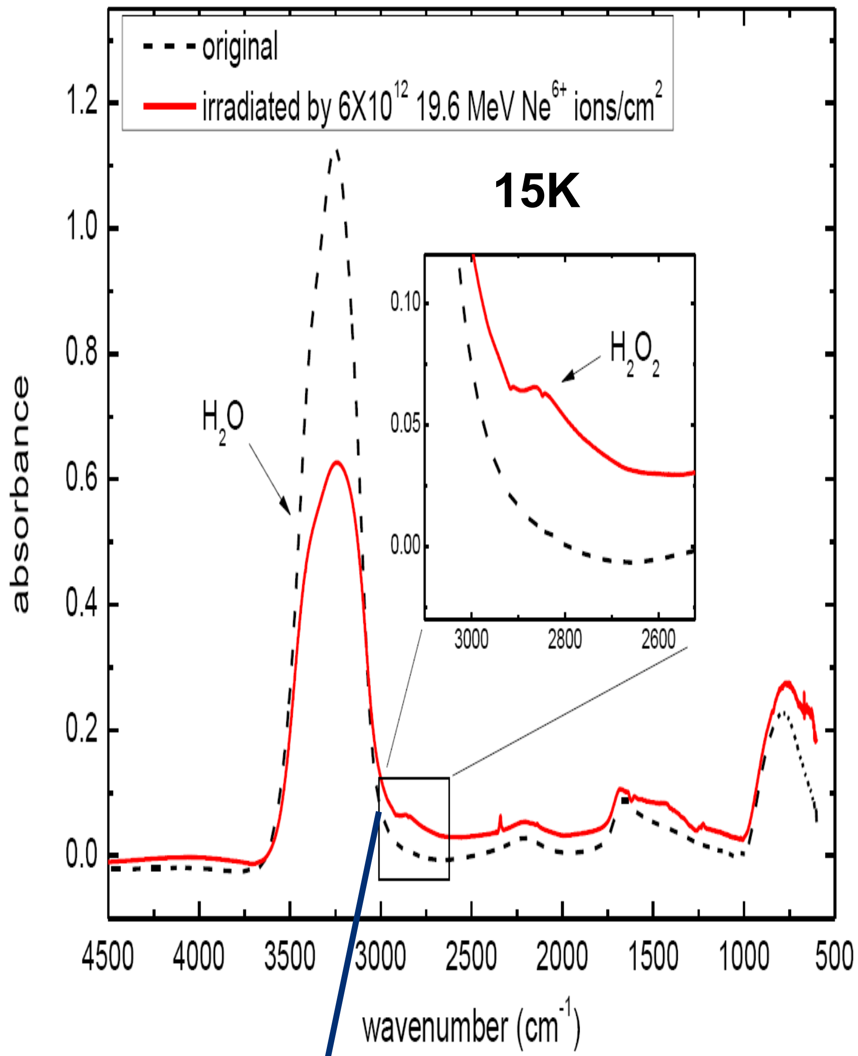
S. Pilling et al.
Astronomy & Astrophysics
509 (2010) A87

B

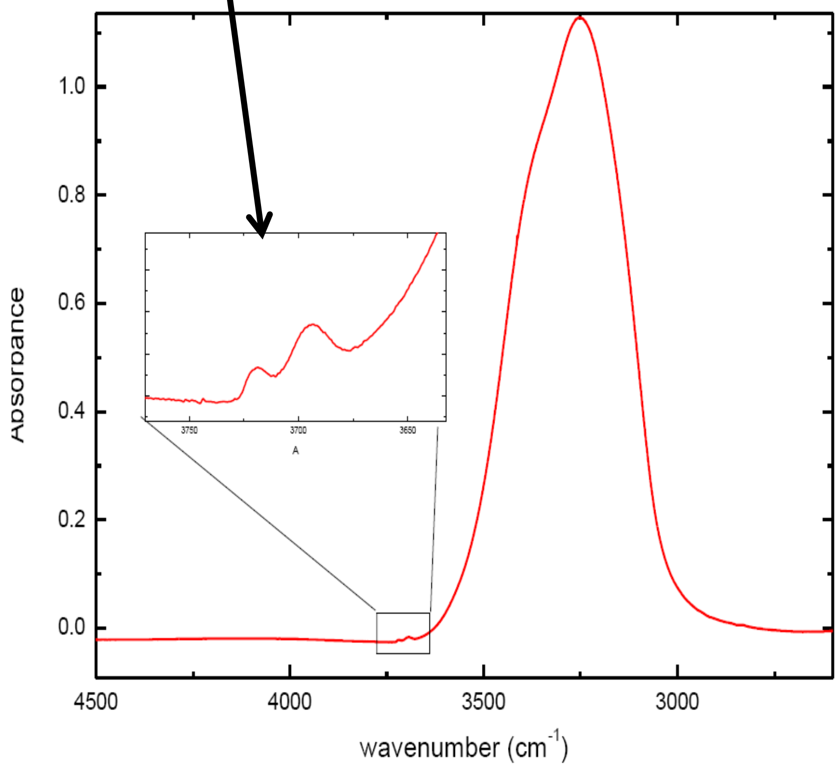
Water ice:
Compaction
and
Amorphization

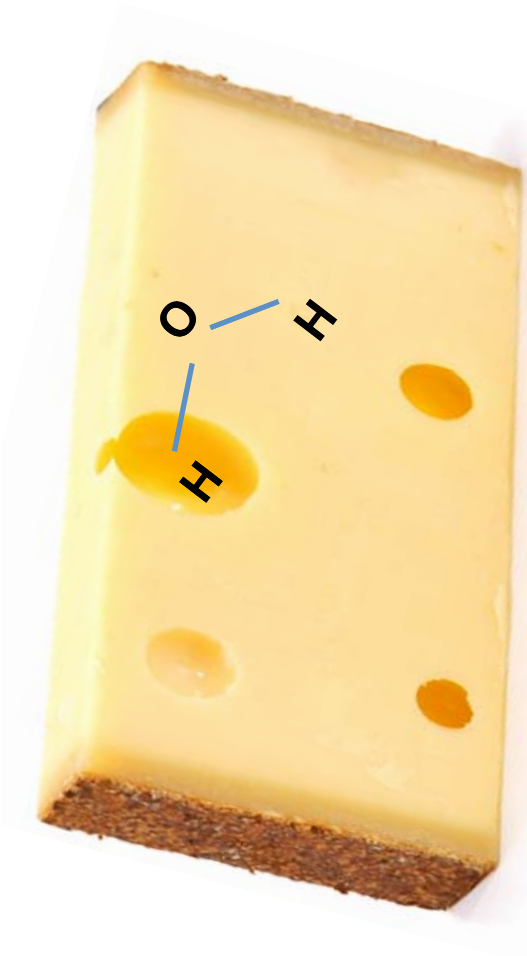
The most abundant molecule in interstellar ices:
Water H_2O

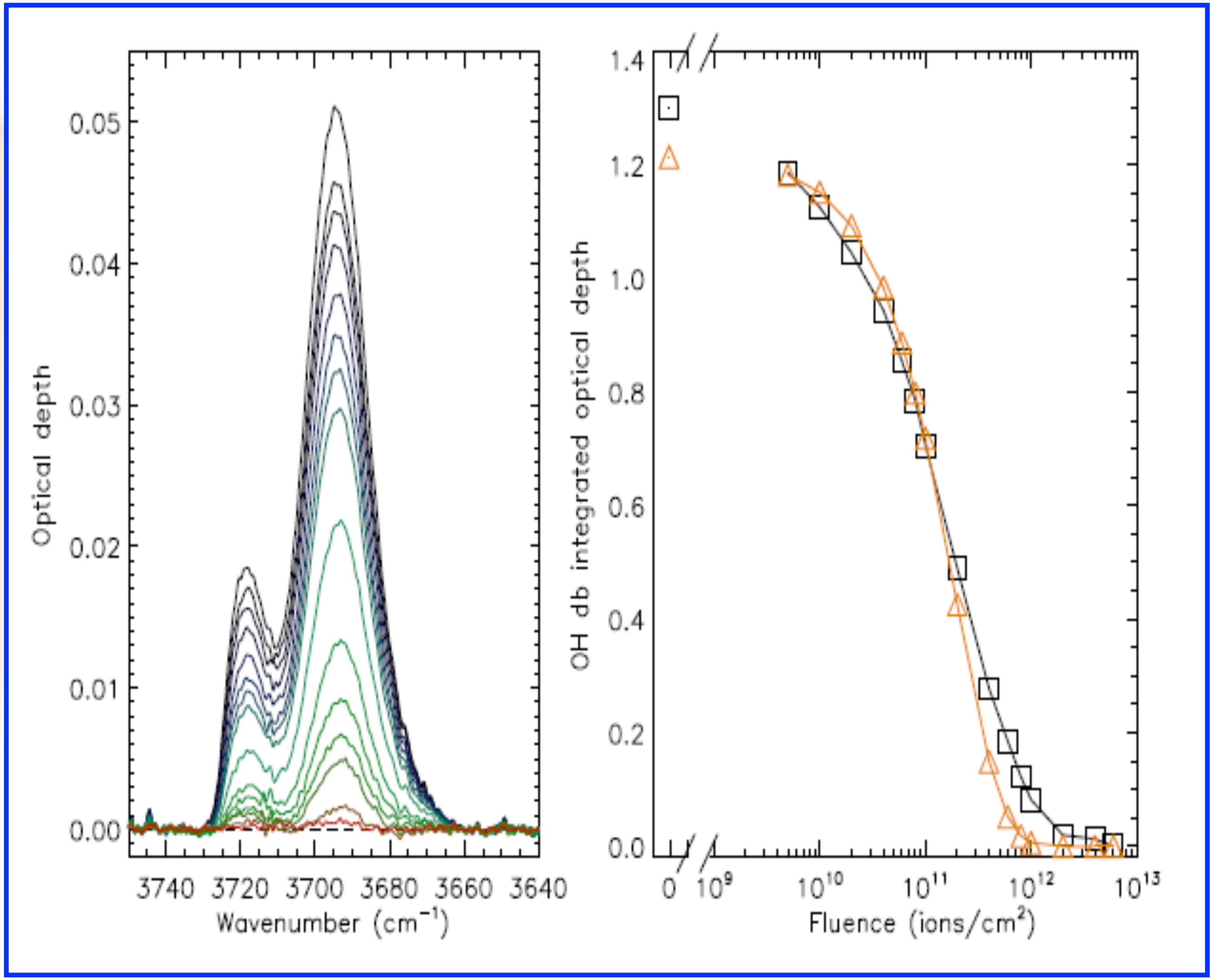
Porosity:
OH dangling bonds

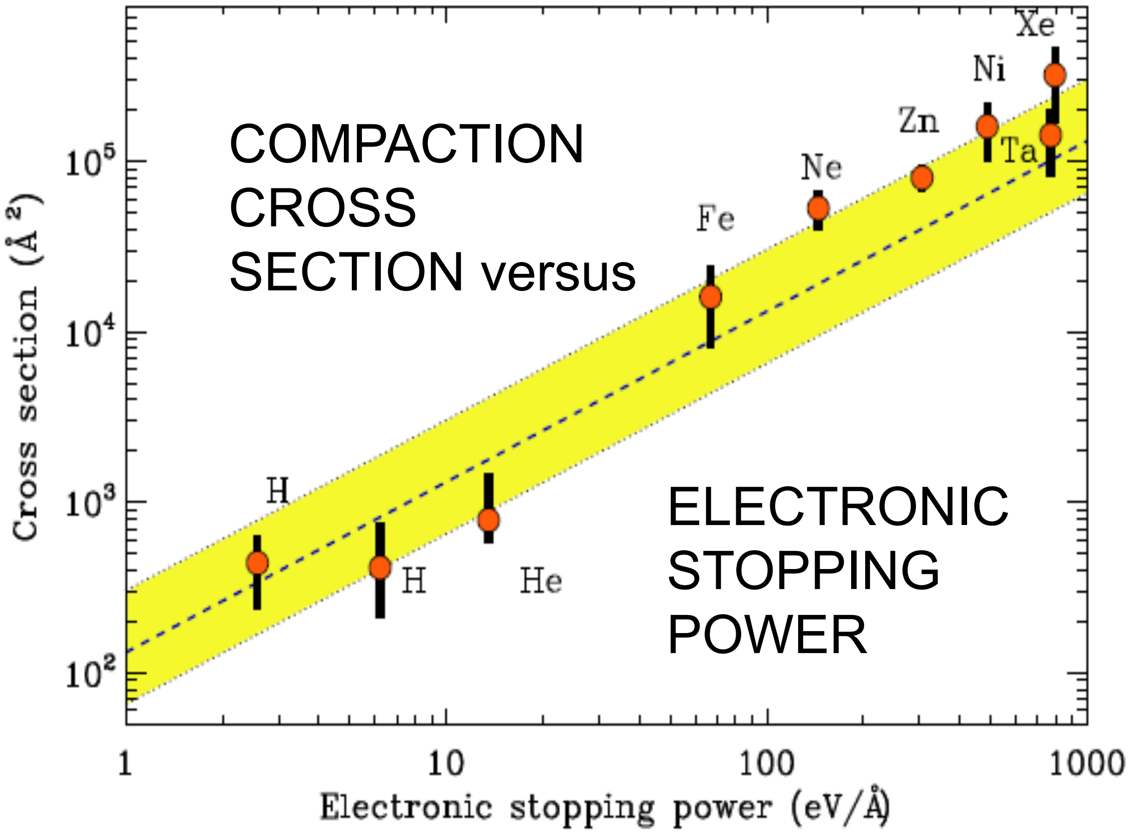


Irradiation of H_2O ice:
formation of H_2O_2




O
H
H


Optical depth
Wavenumber (cm^{-1})
0.00
0.01
0.02
0.03
0.04
0.05
3740
3720
3700
3680
3660
3640
OH db integrated optical depth
Fluence (ions/cm^2)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
0
10^9
10^{10}
10^{11}
10^{12}
10^{13}



t_{comp} = 1 $\times 10^5$ to 2 $\times 10^6$ years

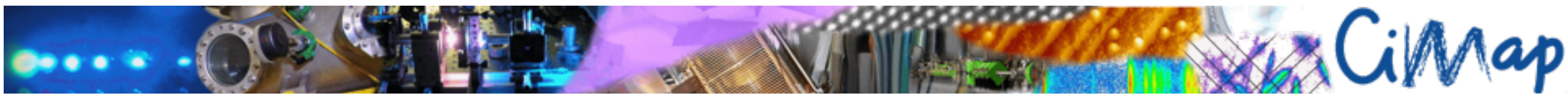
Indeed **no**
OH dangling bonds
observed by
ISO in ISM

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

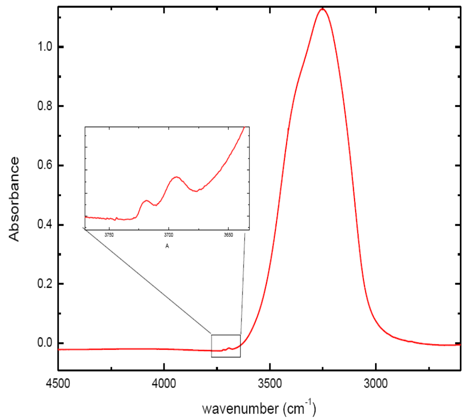
E. Dartois, J.J. Ding, A.L.F. de Barros, P. Boduch, R. Brunetto, M. Chabot, A. Domaracka, M. Godard, X.Y. Lv, C.F. Mejia Guaman, T. Pino, H. Rothard, E.F. da Silveira, J.C. Thomas

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

Astronomy & Astrophysics 557 (2013) A97



Porosity: OH dangling bonds



E. Dartois et al., Astronomy & Astrophysics <u>557</u> (2013) A97

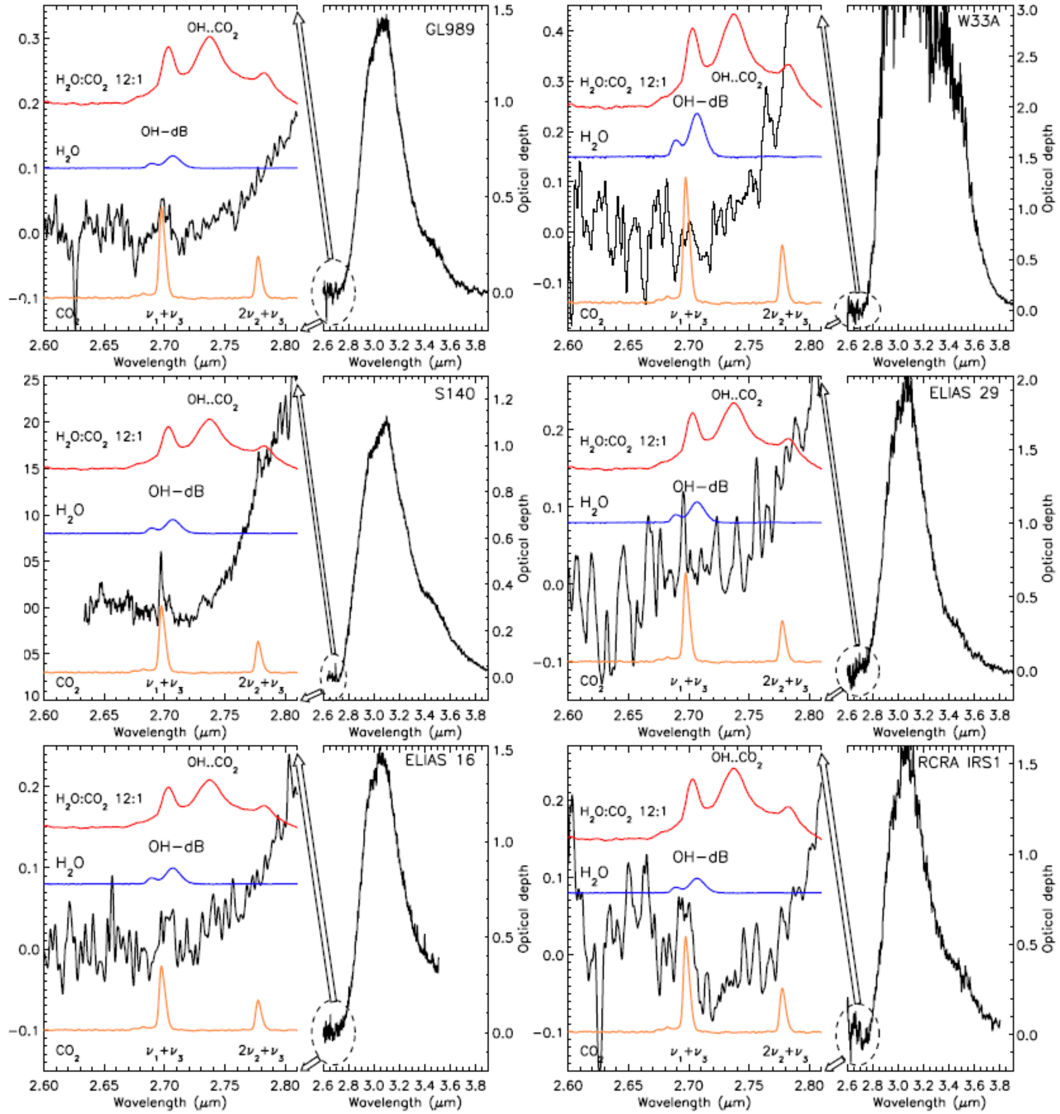
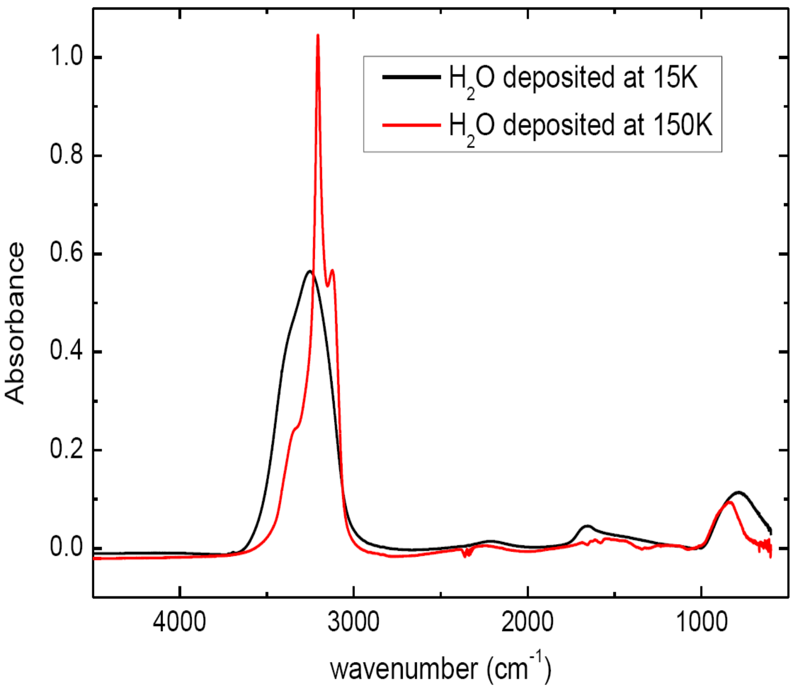
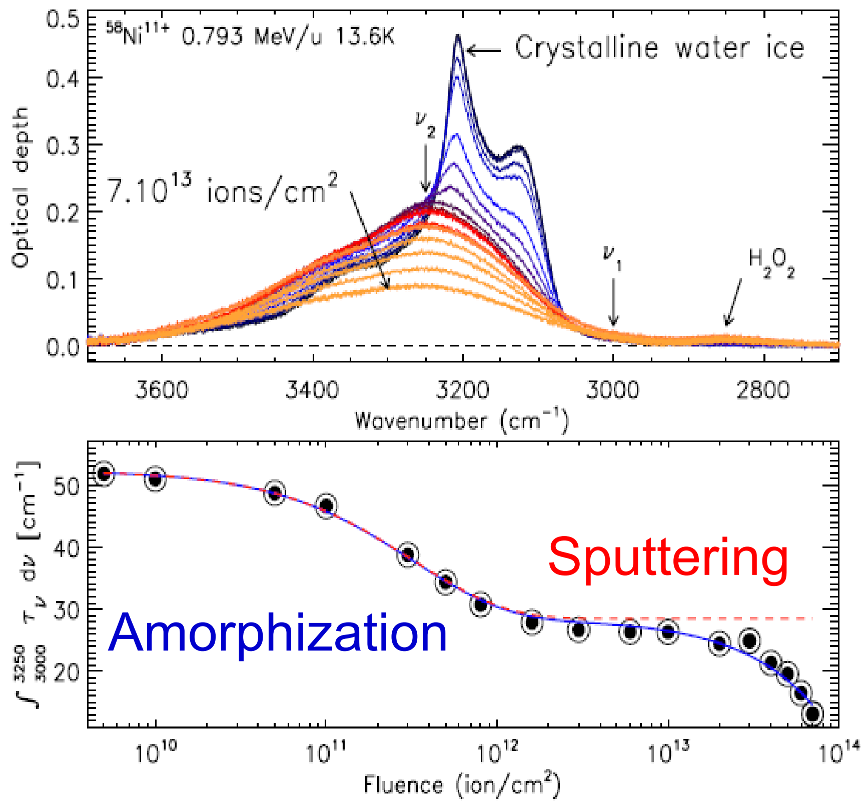


Fig. 4. Young stellar objects spectra measured with the Short Wavelength Spectrometer onboard the Infrared Space Observatory, compared to various laboratory ice mixtures recorded spectra. The optical depth astrophysical spectra are obtained from a continuum baseline subtraction. A close up on the dangling bond region is plotted in the left part of each panel. These spectra are compared to laboratory spectra of pure H_2O and CO_2 ice, and a H_2O:CO_2 12:1 mixture. The CO_2 and a H_2O:CO_2 laboratory spectra are normalised to the CO_2 column density observed in the mid-IR. The H_2O dangling bond spectrum was normalized with the relation discussed in the preceeding section, assuming a porosity of 0.2, and integrating the OH stretching mode. See text for details.

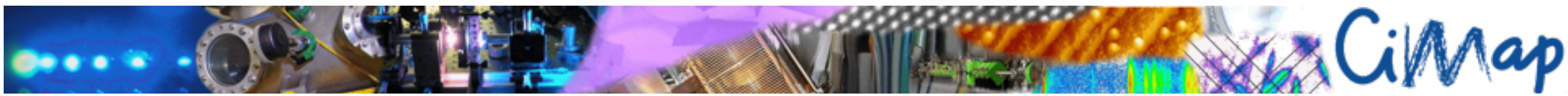




Ion irradiation
3 times more efficient
for compaction than
for amorphization

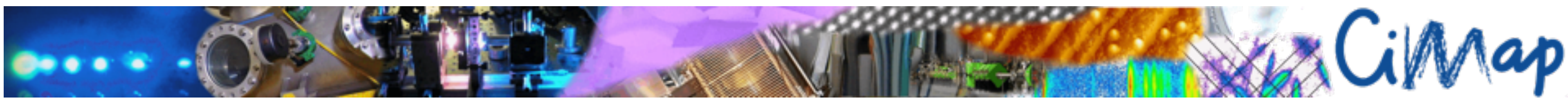
water ice resistant to
phase transition

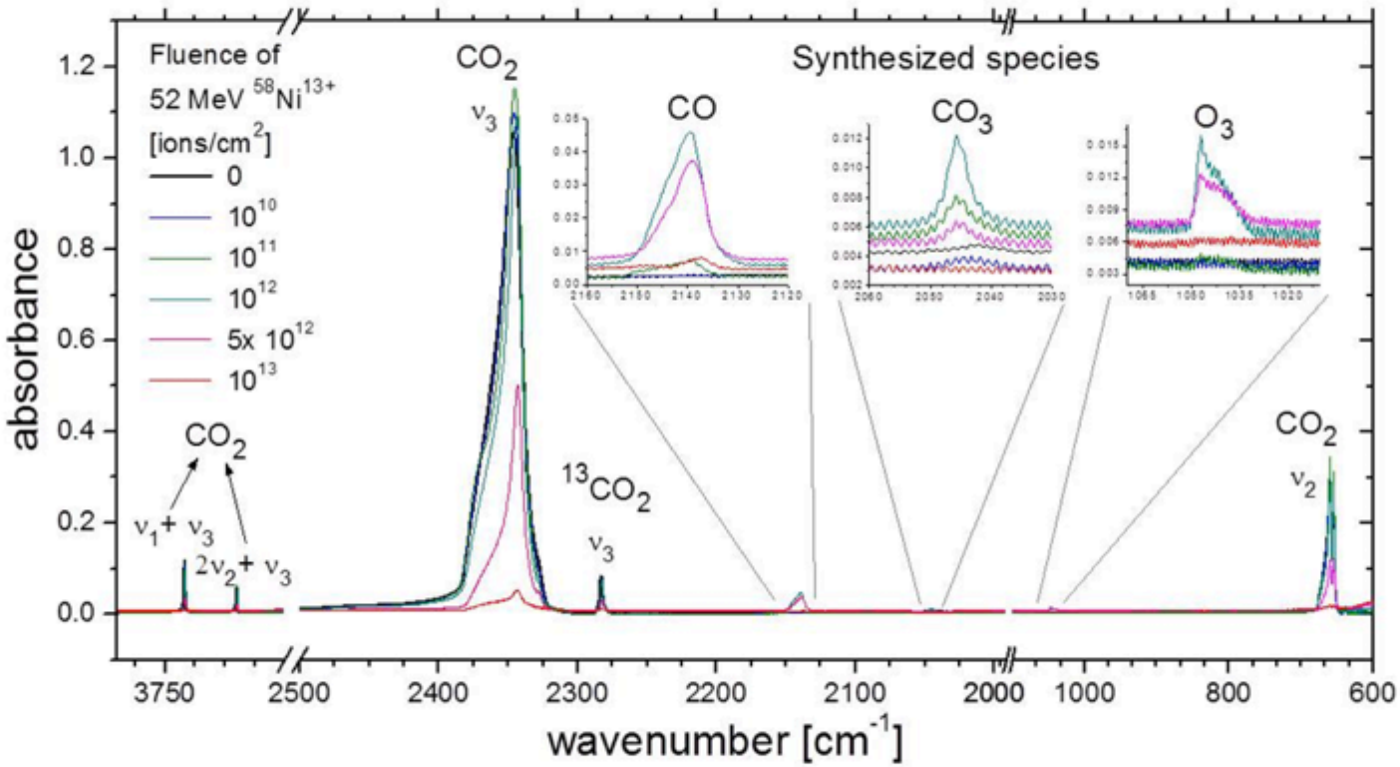
E. Dartois, B. Augé, P. Boduch, R. Brunetto, M. Chabot,
A. Domaracka, J.J. Ding, O. Kamalou, X.Y .Lv,
B. H. Rothard, E.F. da Silveira, J.C. Thomas
Heavy ion irradiation of crystalline water ice -Cosmic ray amorphization cross-section and sputtering yield
Astronomy & Astrophysics 576 (2015) A126



C

Radiolysis of
CO and CO_2 in
dense molecular Clouds:
UV versus Cosmic Rays.





CO_2

@ GSI + GANIL

projectiles:

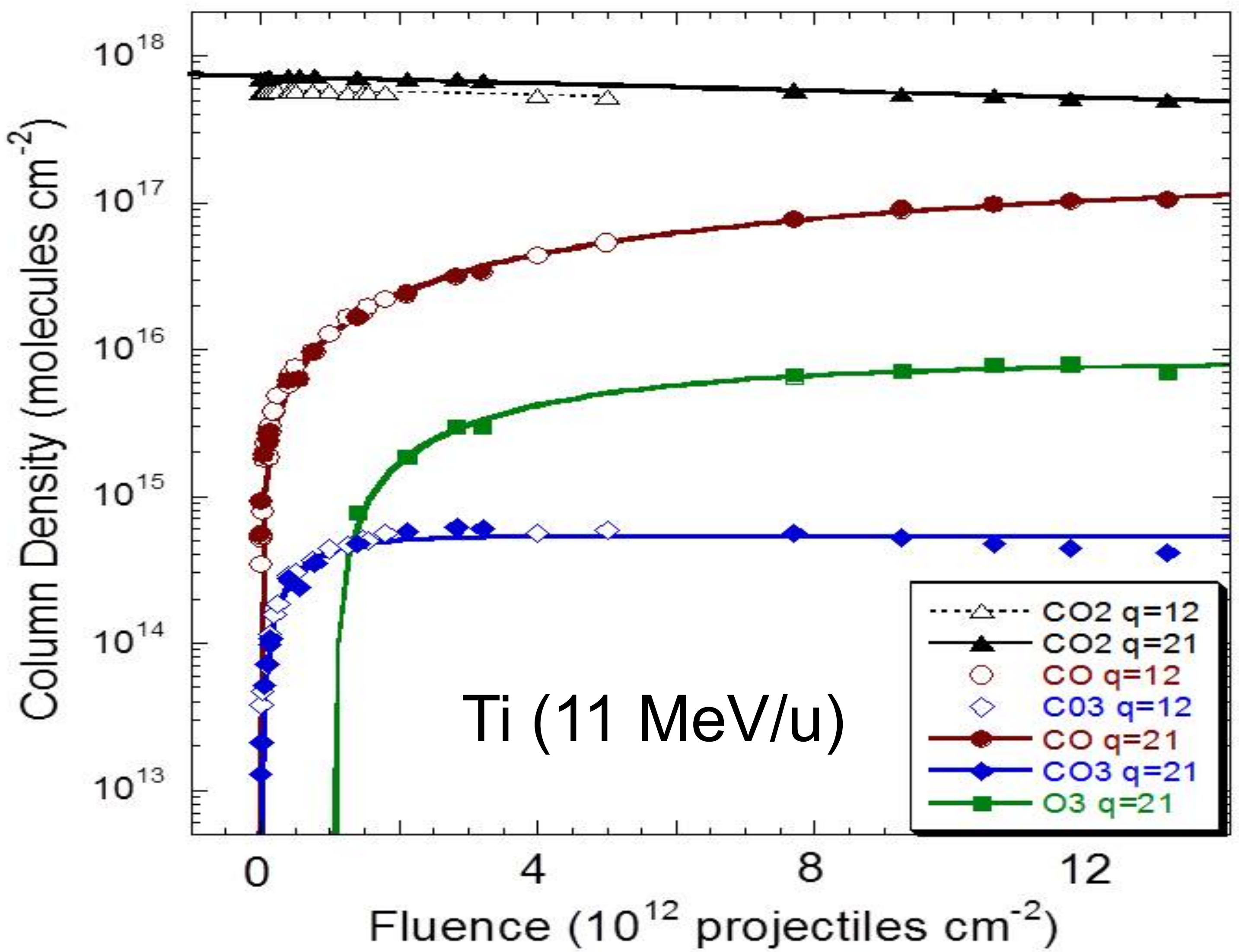
Ni

Ti

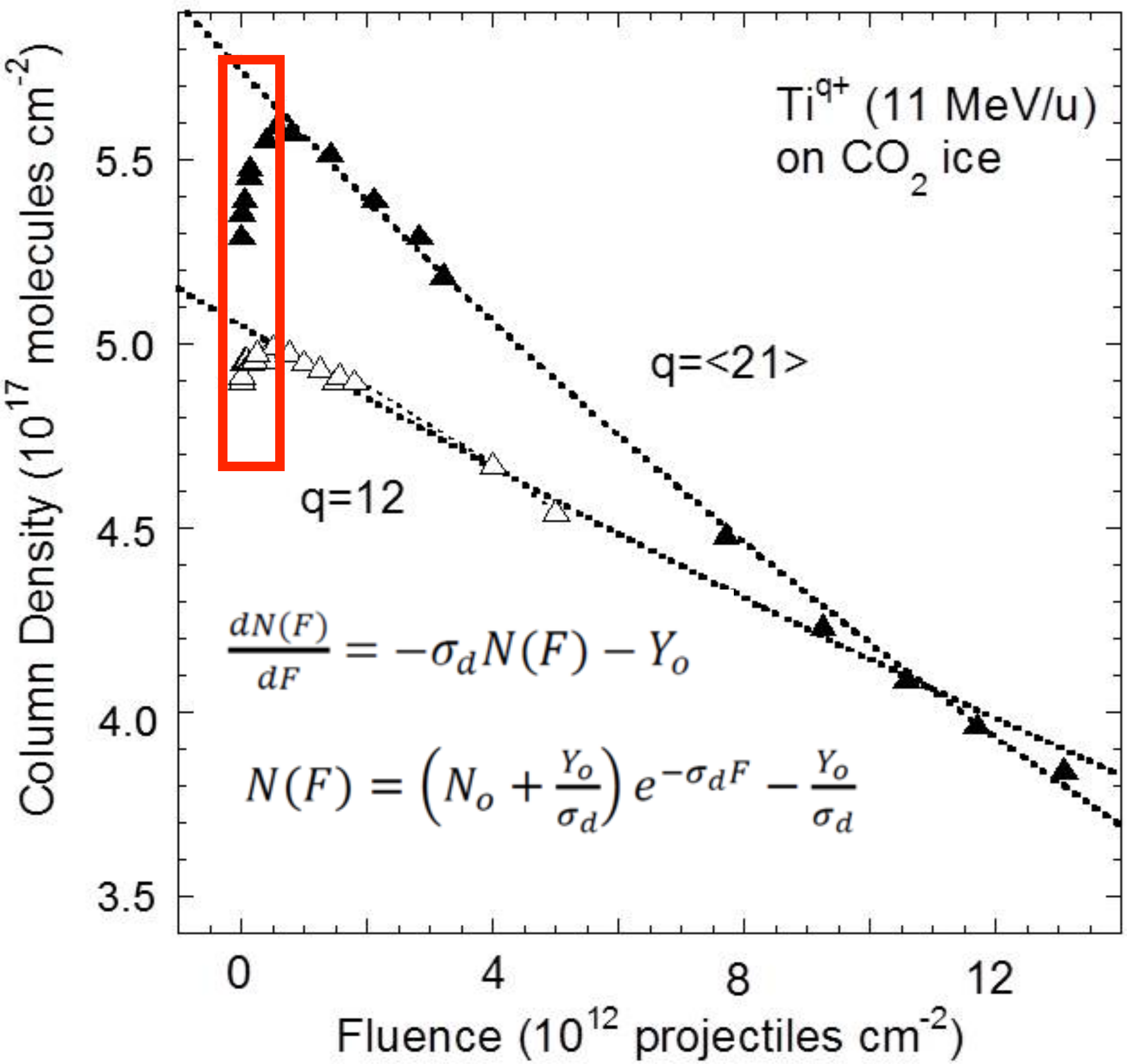
Xe

Materialforschung

Projectile fluence dependence



Projectile fluence dependence



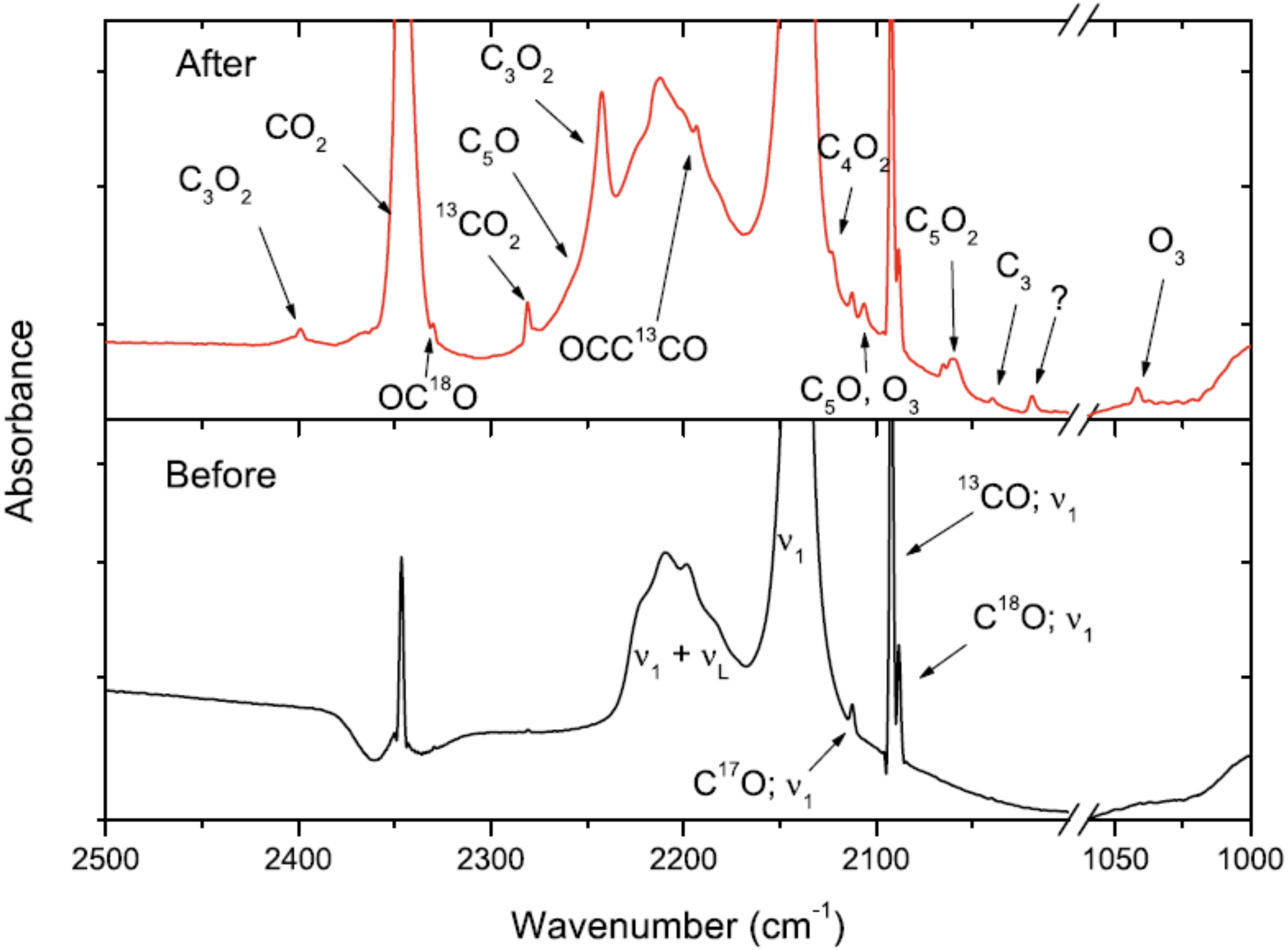
CO_2:
Compaction*
destruction
(fragmentation)
sputtering

Formation of
CO, O_3, CO_3

*C.F. Mejía, A.L.F. de Barros, E. Seperuelo Duarte, E.F. da Silveira, E. Dartois, A. Domaracka, H. Rothard, P. Boduch, **Compaction of porous ices rich in water by swift heavy ions** Icarus 250 (2015) 222

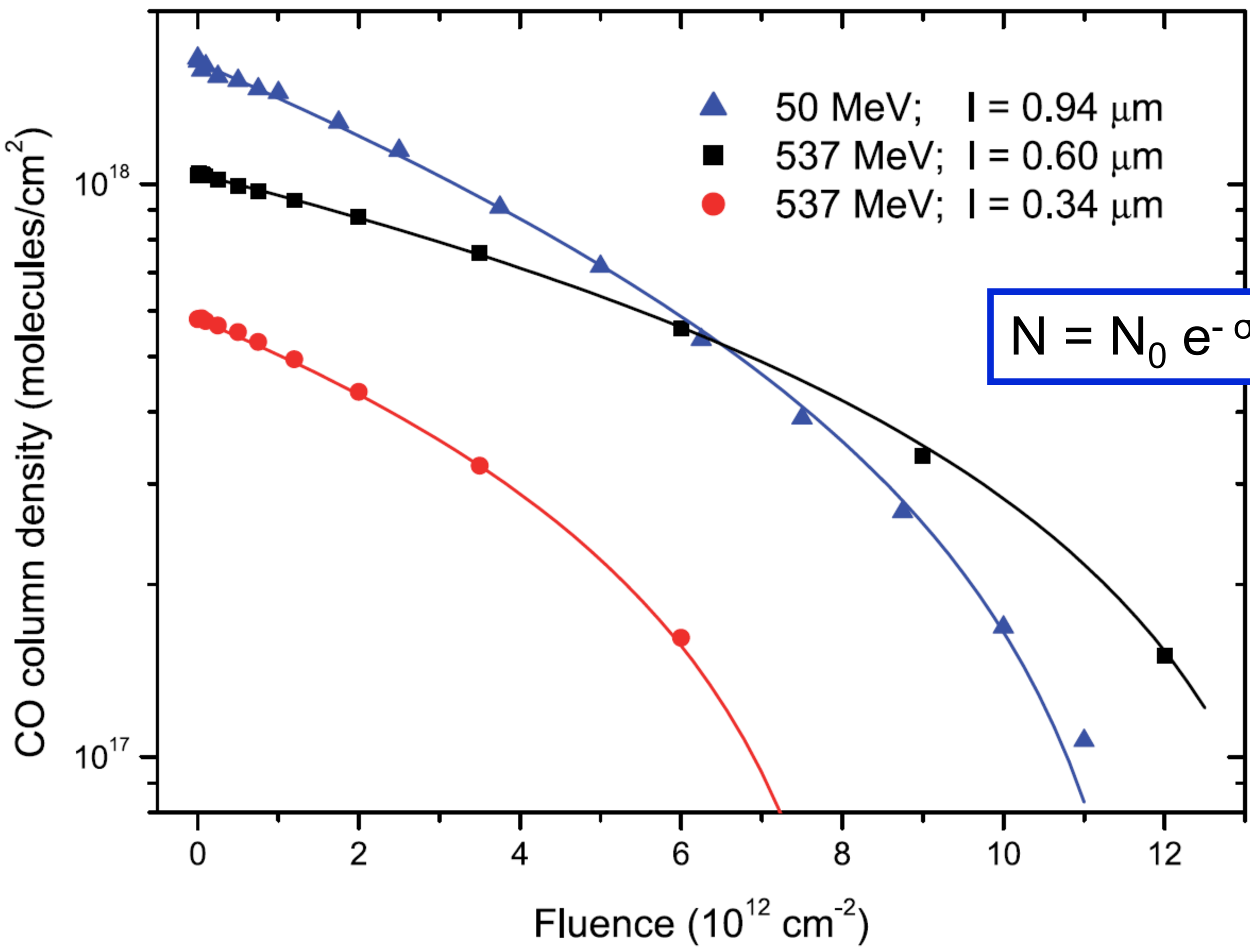
Another example: CO ice

(the second most abundant molecule in space ices after H_2O)



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

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



$$N = N_0\, e^{-\sigma_d F} - (\mathbf{Y}/\sigma_d)\,(1 - e^{-\sigma_d F})$$

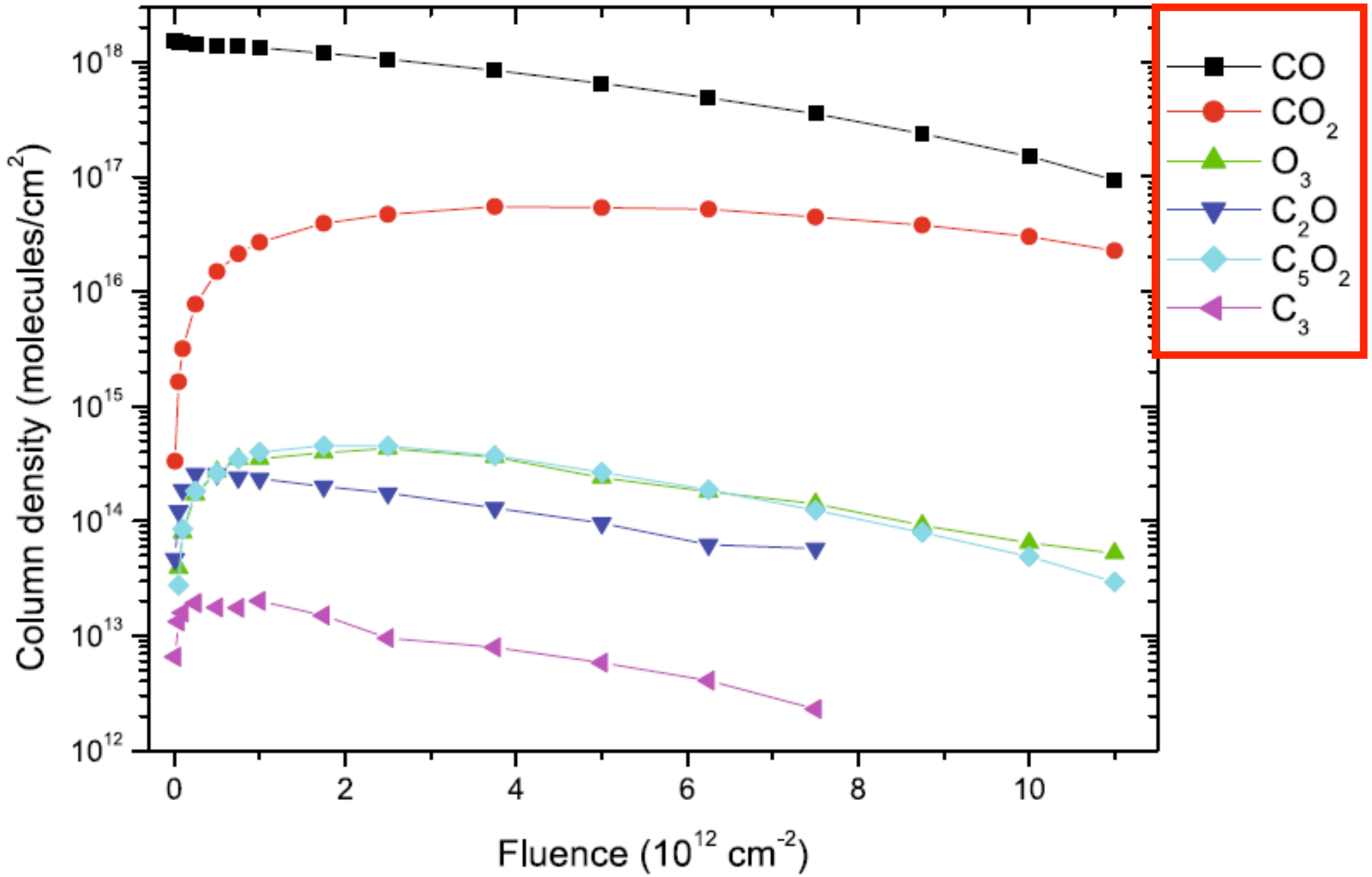
deduced quantities:

Destruction Cross Section $\boldsymbol{\sigma_d}$

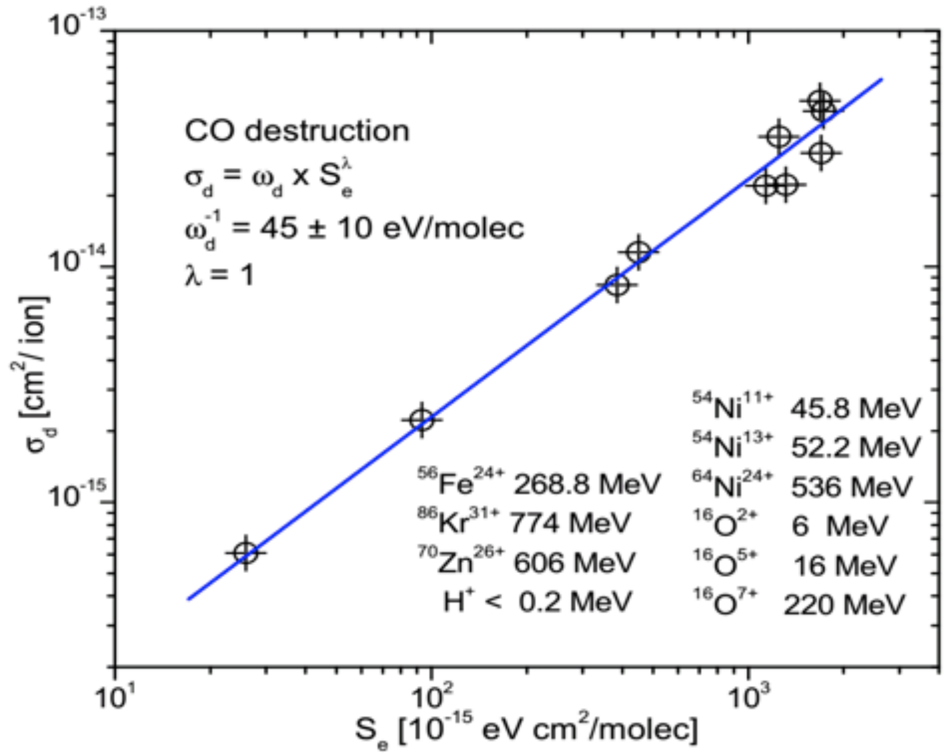
Sputtering Yield Y

Formation cross section for daugther molecules $\boldsymbol{\sigma_f}$

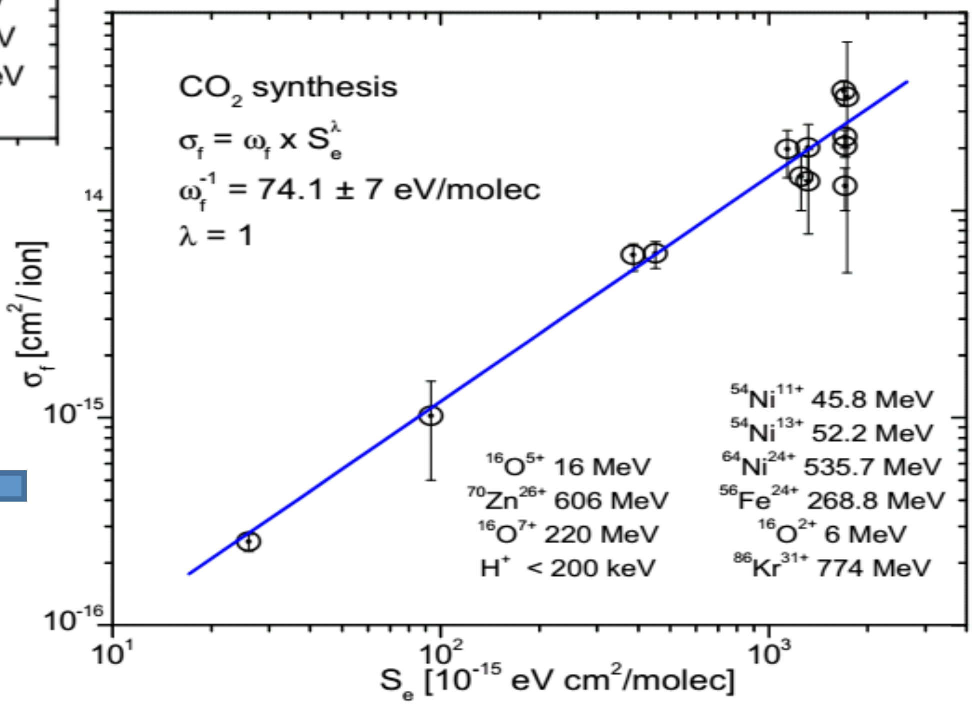
CO ice: formation of new molecular species



Column density of CO and molecules produced as a function of fluence of 50 MeV Ni ions.

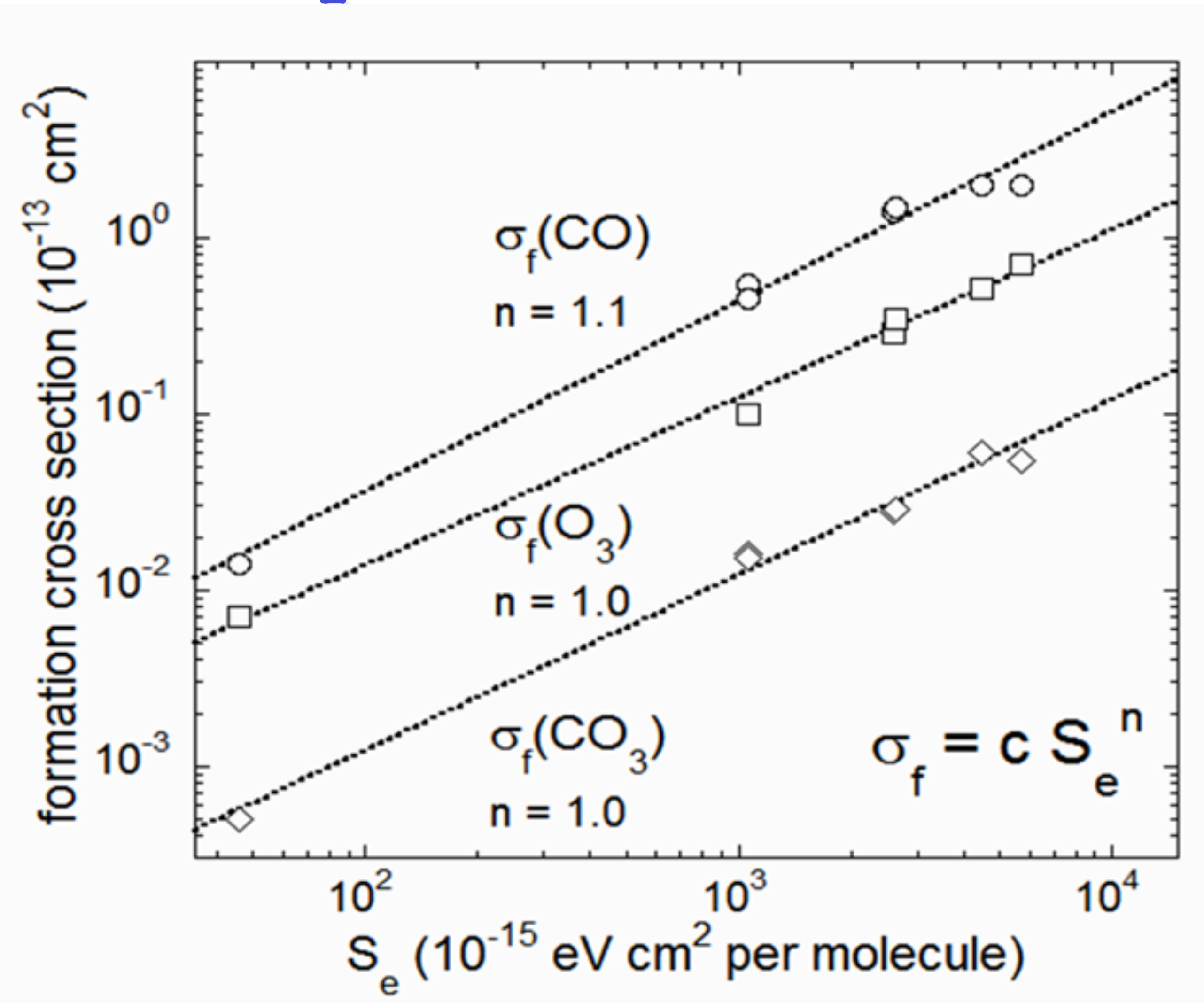


CO destruction cross section proportional to S_e



CO_2 formation cross section proportional to S_e

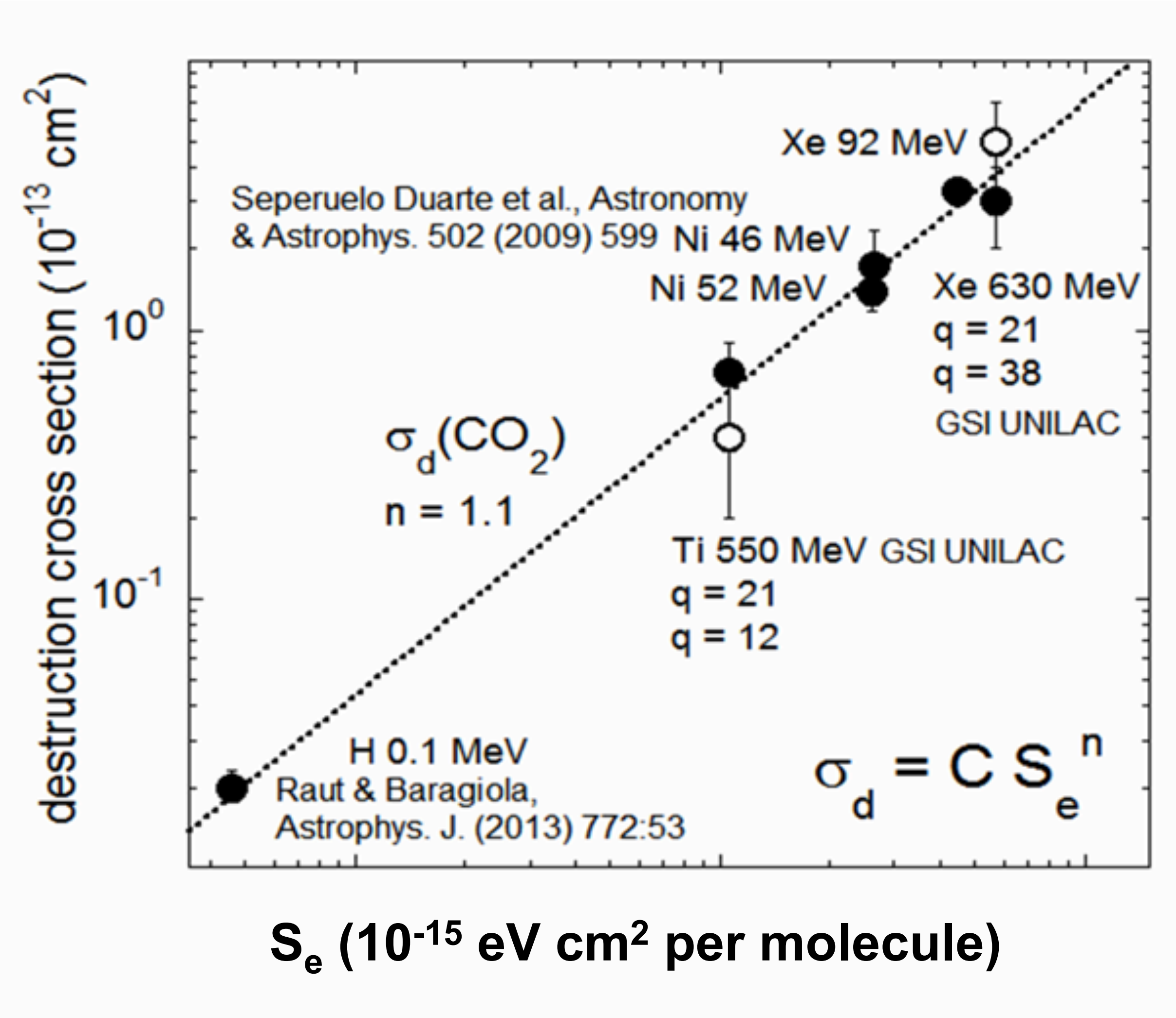
CO_2: Formation cross sections



again:

formation cross sections proportional to S_e

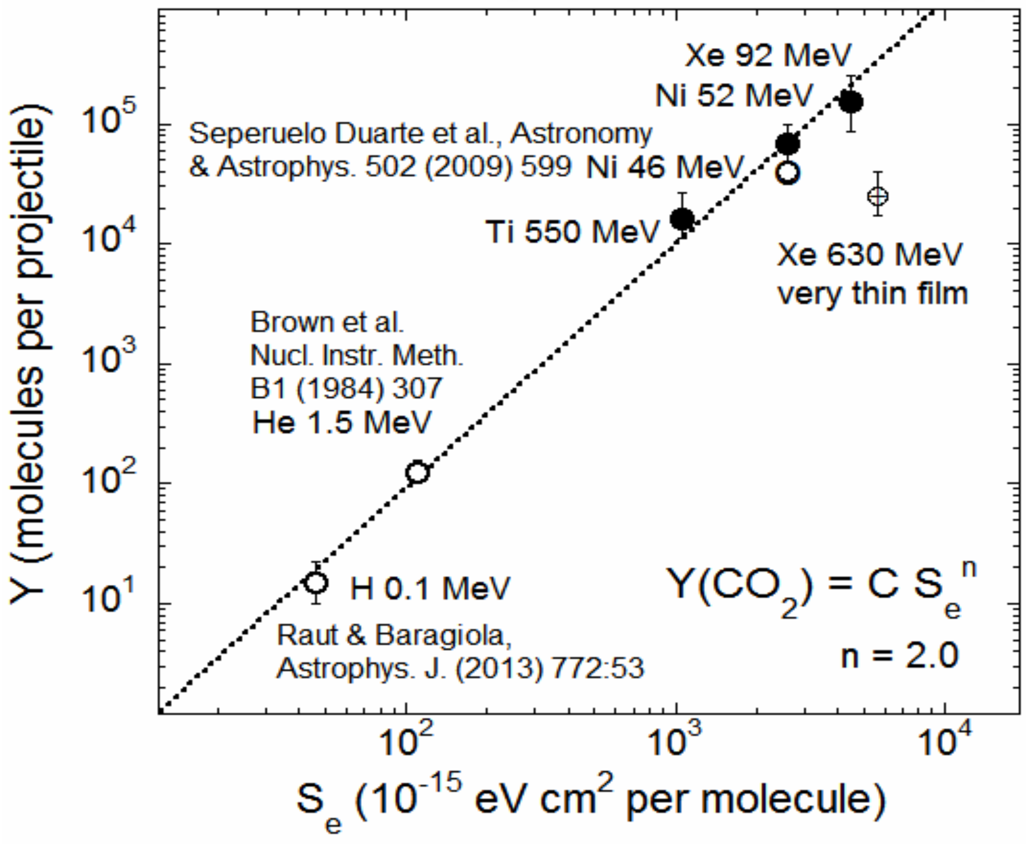
CO_2: Destruction cross sections



again:

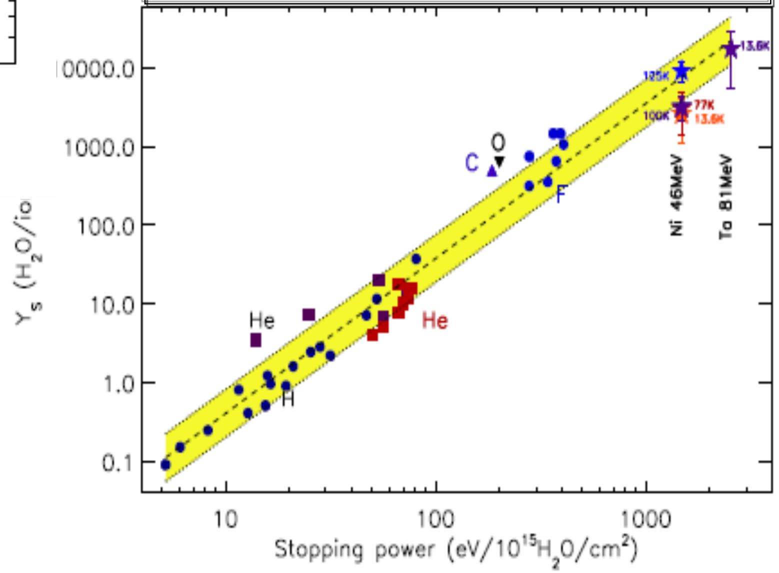
destruction cross section proportional to S_e

CO_2: Sputtering yield

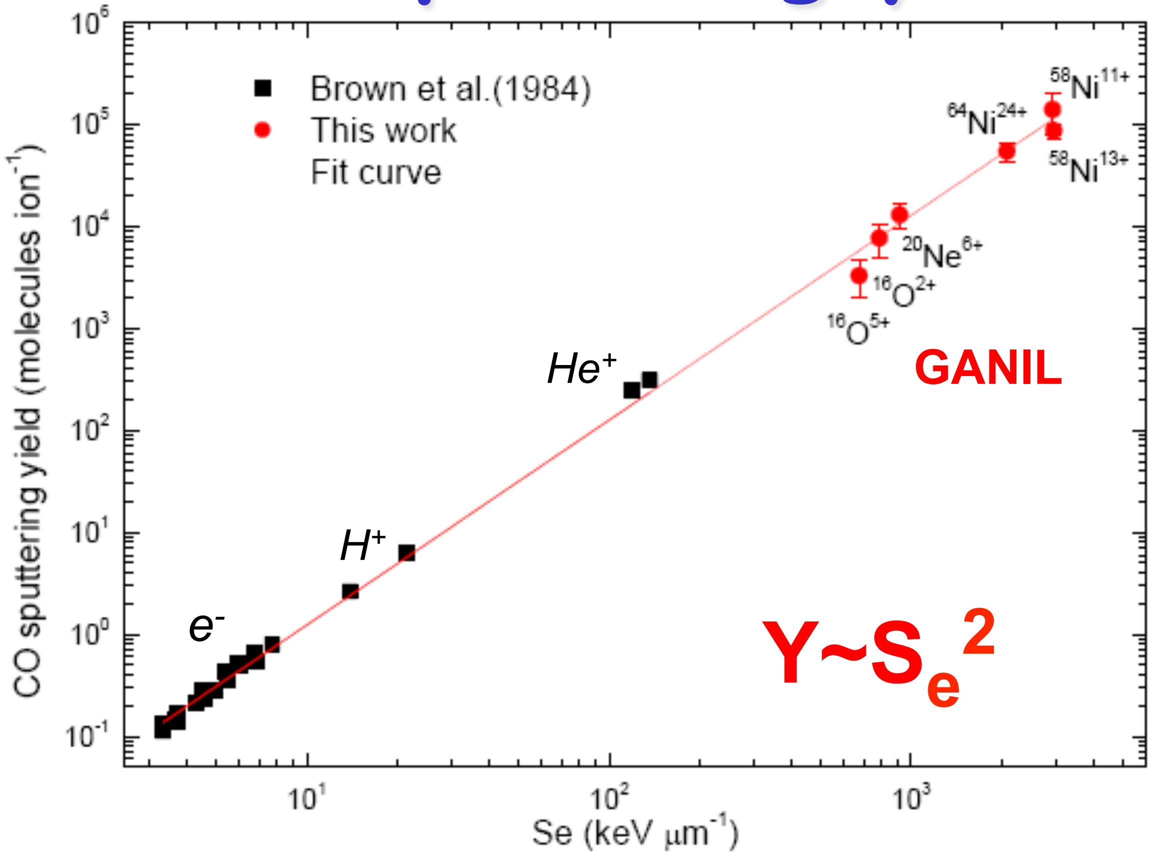


Y scales with $S_e{}^2$!

also true for **H_2O**!
estimation for
galactic cosmic rays:
$Y_{GCR}(H_2O) \approx 10\ H_2O/cm^2/s$



CO: Sputtering yield



again:

$Y \sim S_e^2$

(universal ...)

$S_e \sim Z_P^2$

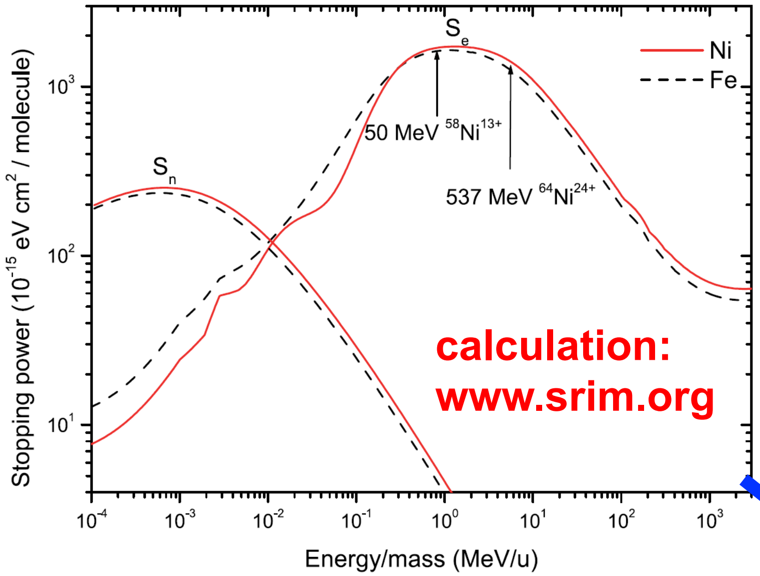
$Y \sim Z_P^4$

very strong dependence!

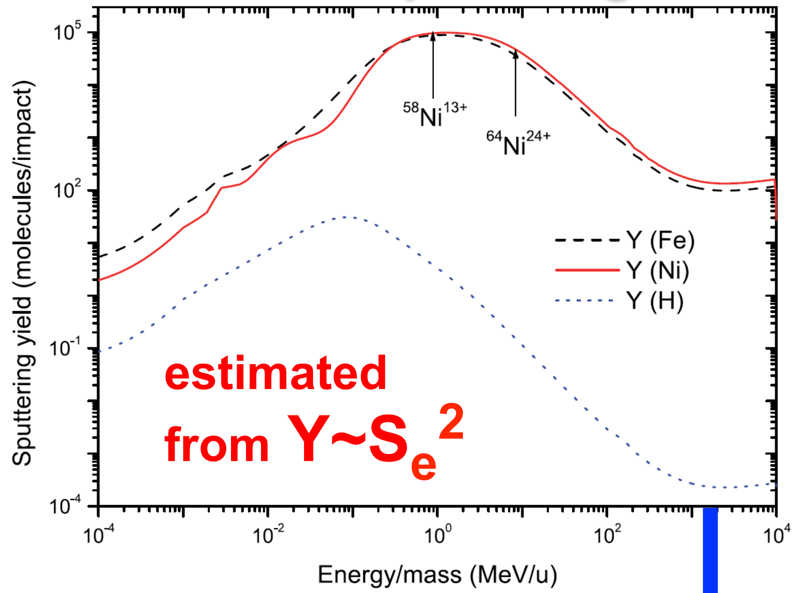
W.L. Brown, W.M. Augustyniak, K.J. Marcantonio, E.H. Simmons, J.W. Boring, R.E. Johnson, C.T. Reimann, Nucl. Instr. Meth. B1 (1984) 307

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

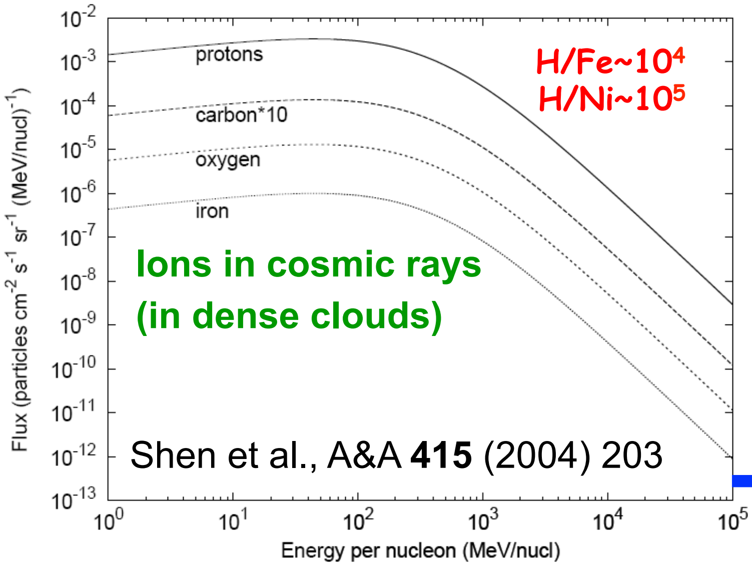
Electronic Energy Loss in CO

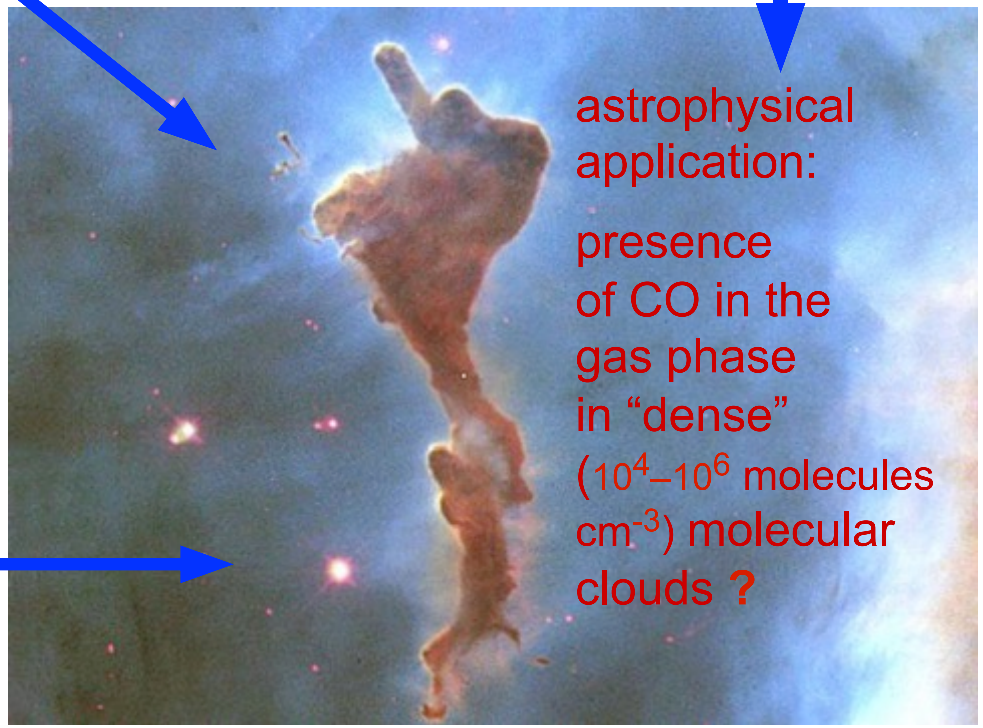


CO Sputtering Yield

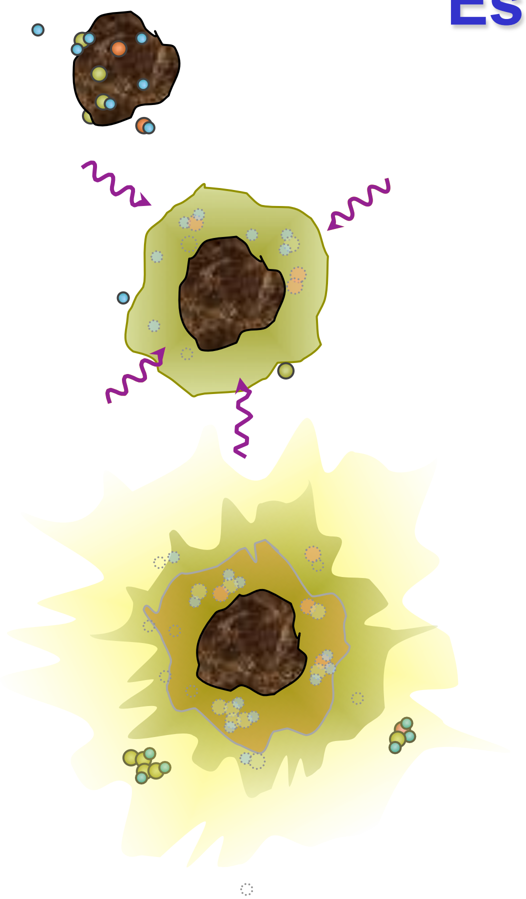


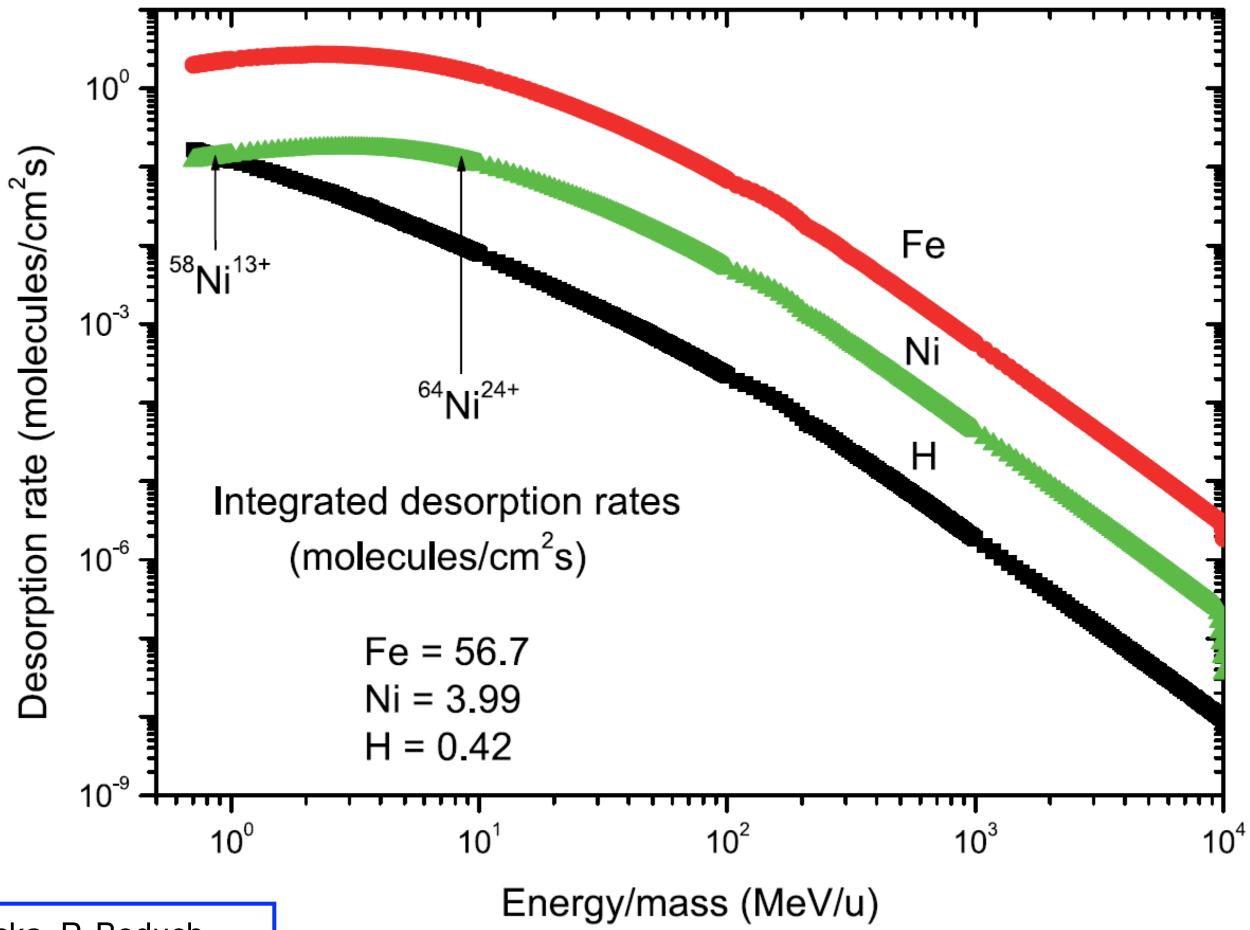
Heavy ion Abundance in space





Estimated ion induced CO Desorption Yield

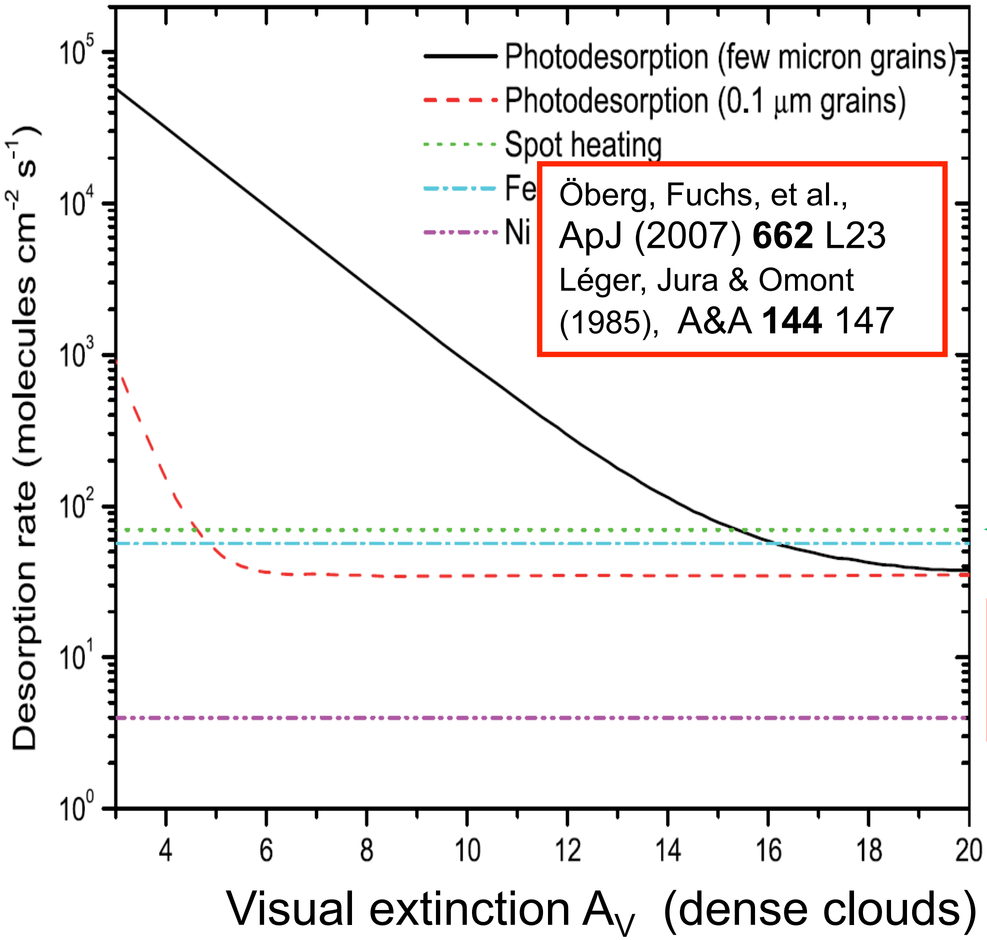




E. Seperuelo Duarte, A. Domaracka, P. Boduch,
H. Rothard,E. Dartois, E.F. da Silveira
Laboratory simulation of heavy ion cosmic ray interaction with condensed CO
Astronomy & Astrophysics 512 (2010) A71

Desorption rate of CO as a function of visual extinction

pentration depth dependence in dense molecular clouds



Heavy Ions

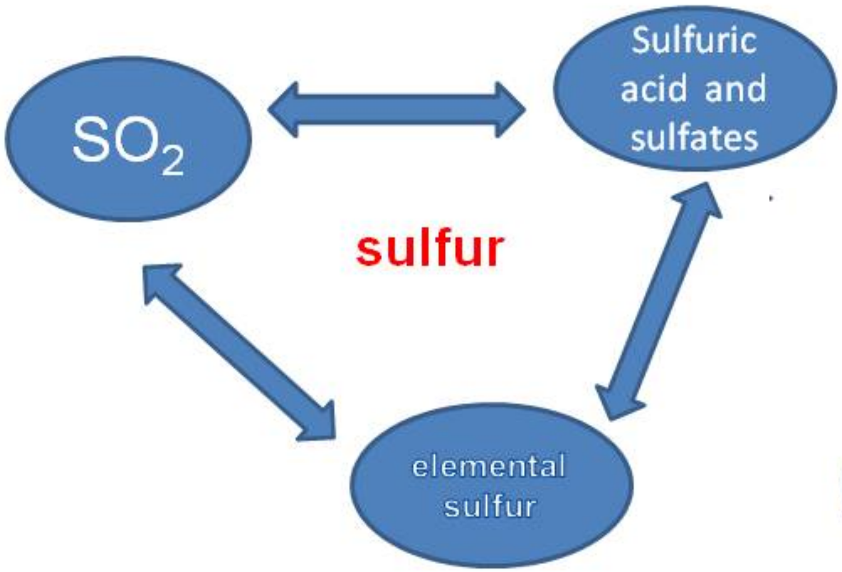
UV photons

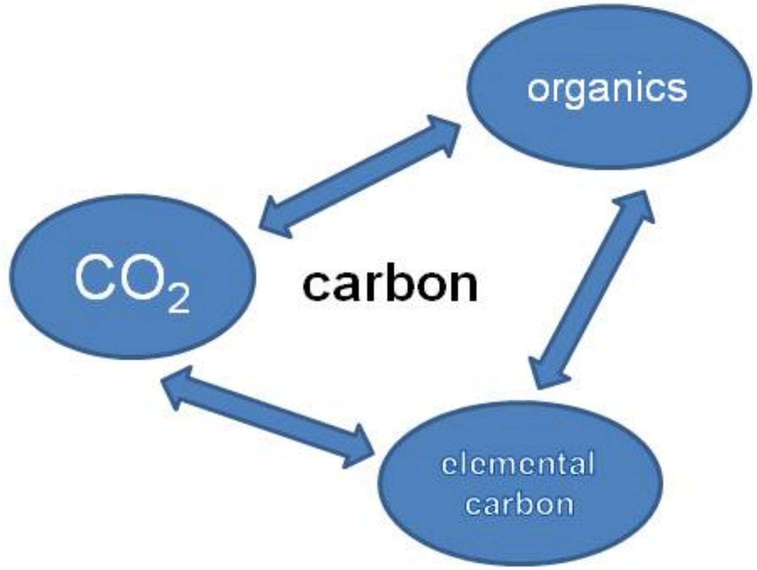
E. Seperuelo Duarte, A. Domaracka, P. Boduch, H. Rothard, E. Dartois, E.F. da Silveira
Astronomy & Astrophysics 512 (2010) A71

D

Galilean Moons, Jupiter’s Magnetosphere, and Sulfur and Carbon cycles.

cycles





endogenic versus exogenic origin ?
processing ?
who is the first ?

Is CO_2 produced by C^{n+} ion implantation **?**

Are SO_2 or H_2SO_4 produced

by S^{n+} ion implantation **?**

Jupiter, NASA's spacecraft *GALILEO*, and the Galilean Moons Io, Europa, Ganymede, Callisto

Io: SO_2 ice dominant

Europa, Callisto, Ganymede: H_2O ice dominant

Europa: significant quantities of magnesium, sodium sulfate Na_2SO_4, carbonate hydrates

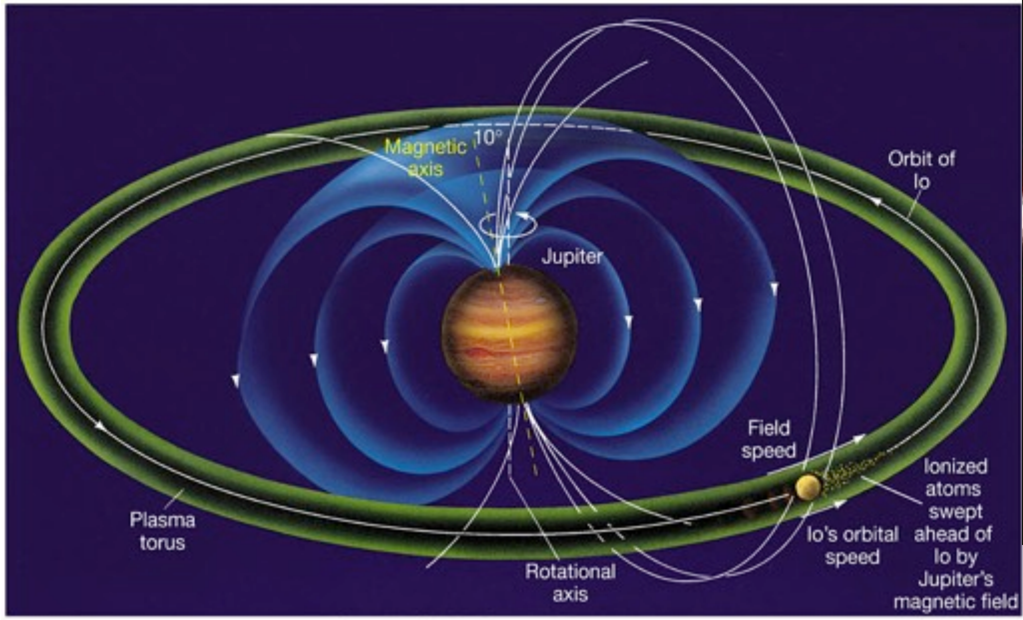
Other absorption features and prime candidates:

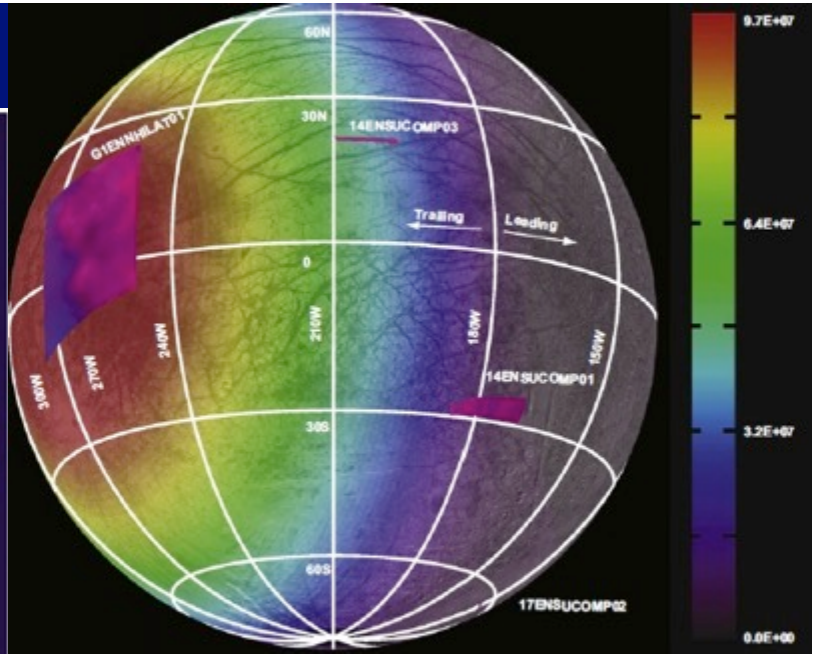
3.4 μm		(~2940 cm^{-1})	C-H
3.5	"	(~2857 cm^{-1})	H_2O_2
3.88	"	(~2580 cm^{-1})	S-H, H_2CO_3
4.05	"	(~2470 cm^{-1})	SO_2
4.25	"	(~2350 cm^{-1})	CO_2
4.57	"	(~2190 cm^{-1})	CN

Open question: are these species **native** from the satellites or **synthesized** by **exogenic processes** e.g. ion implantation **?**

JUICE **2022 - 2033**
ESA Cosmic Vision

The Jovian Magnetosphere

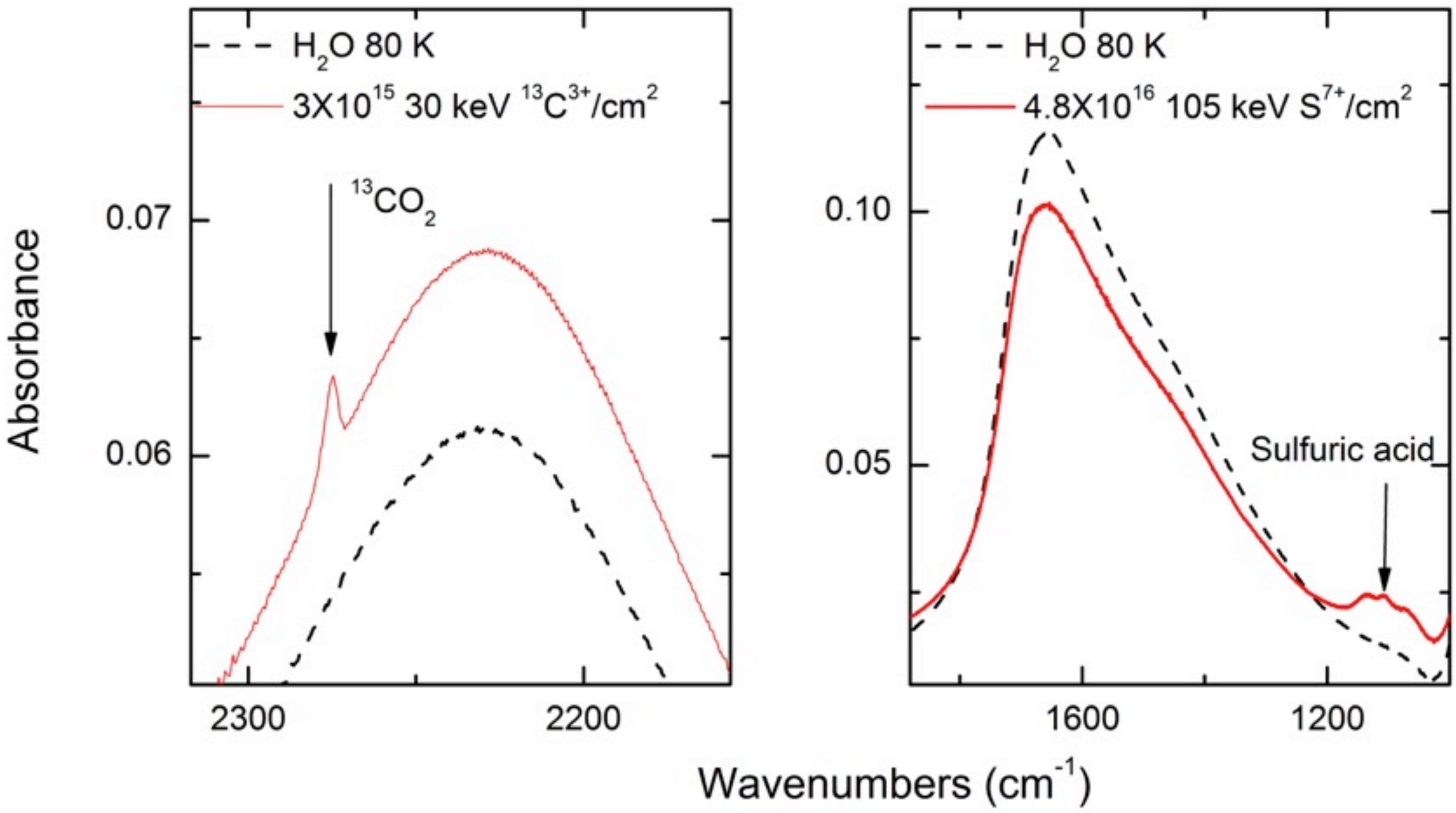




E U R O P A : Sulfur Ion Flux

J.B. Dalton III, T. Cassidy, C. Paranicas,
J.H. Shirley, L.M. Prockter, L.W. Kamp
Planetary and Space Science 77 (2013) 45

What can we do in the laboratory?
we can measure formation yields of carbon dioxide and sulfuric acid!

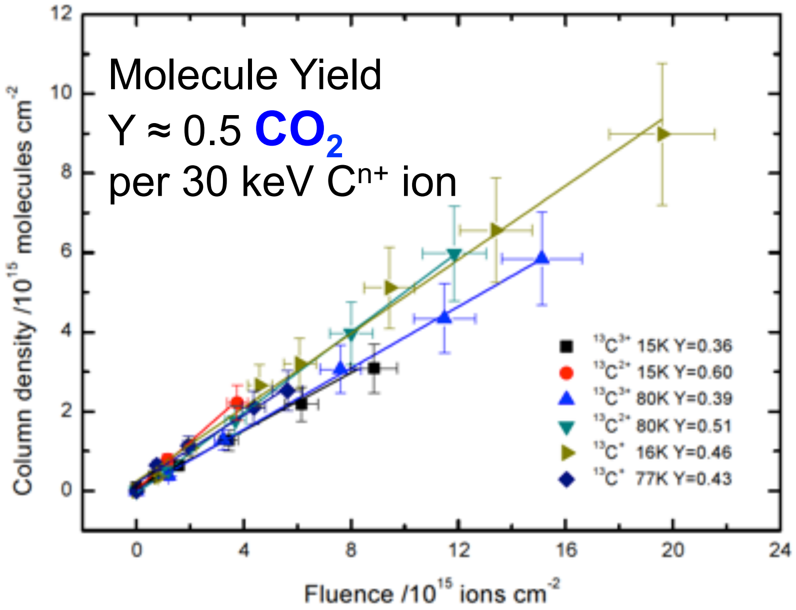


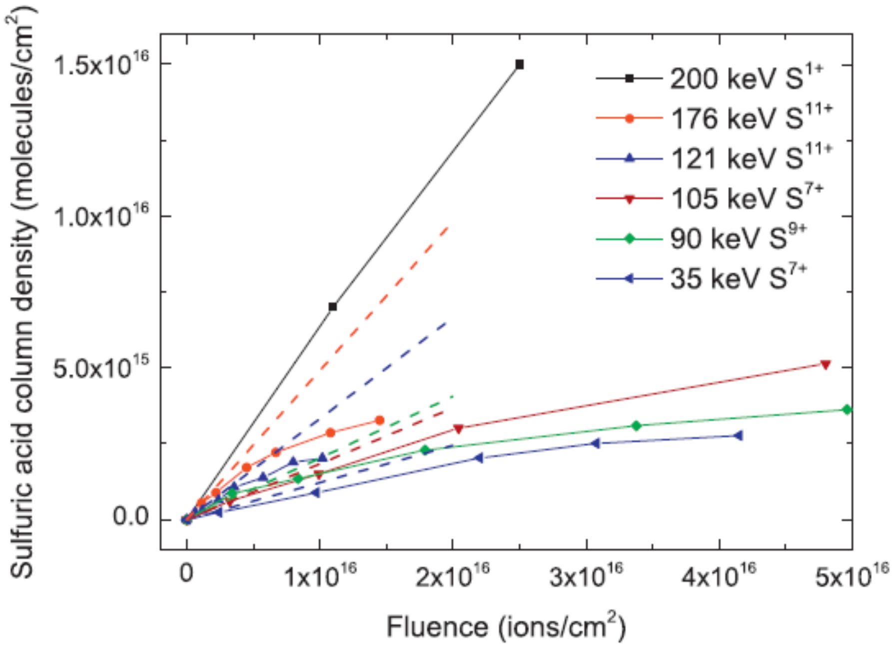
IR spectra of water ice before and after implantation of carbon and sulfur ions

X.Y. Lv, A L F. de Barros, P. Boduch, V. Bordalo, E.F. da Silveira, A. Domaracka, D. Fulvio, C. A.Hunniford, T. Langlinay, N.J. Mason, A.R. W. McCullough, M.E. Palumbo, A.S. Pilling, H. Rothard, G. Strazzulla

Implantation of multiply charged
Carbon Ions in Water Ice

Astronomy & Astrophysics 546 (2012) A81



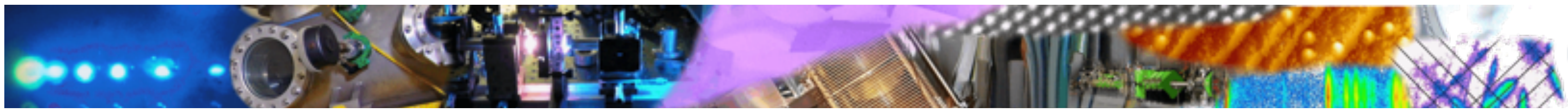


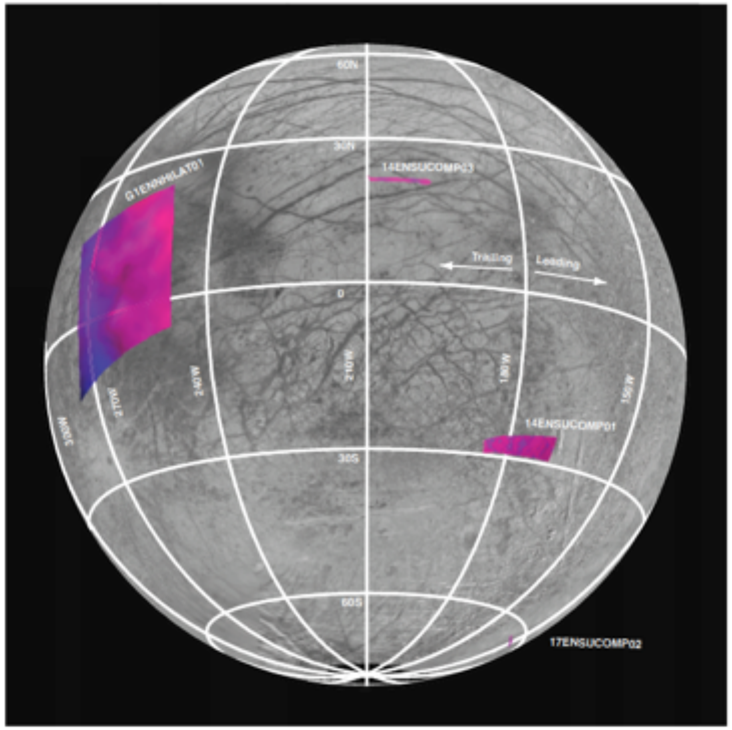
Molecule Yield
Y(H_2SO_4) ≈ 0.12 (35 keV)
≈ 0.64 (200 keV
per S^{n+} ion

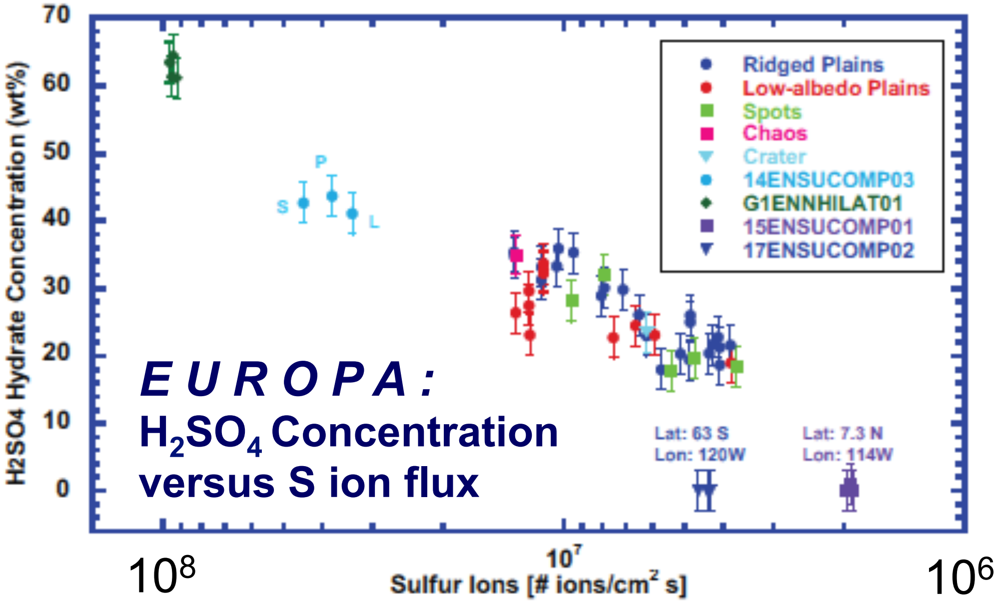
J. J. Ding, P. Boduch, A. Domaracka, S. Guillous, T. Langlinay, X.Y. Lv, M.E. Palumbo, H. Rothard, G. Strazzulla

Implantation of Multiply Charged
Sulfur Ions in Water Ice

Icarus 226 (2013) 860–864







J.B. Dalton III et al.,
Planetary and Space Science 77 (2013) 45:

Correlation of H_2SO_4 hydrate concentration with sulfur ion flux

Ding et al., Icarus 336 (2013) 860:

H_2SO_4 Concentration compatible with measured Molecule Yield from Implantation!

time needed to obtain
3 x 10^{19} molecules/cm^2 by implantation

Flux of S-ions (cm^{-2} s^{-1})	Time (years) Using Y=0.12	Time (years) Using Y=0.64
2×10^6	4×10^6	7×10^5
1×10^8	9×10^4	1.4×10^4

Ding et al., Icarus 336 (2013) 860:

Concentration compatible with measured Molecule Yield from Implantation!

→ **Exogenic production of H_2SO_4 probable**

but ... C implantation in water ice does not explain observed CO_2 concentration.

and ... no evidence (yet) for production of SO_2 or H_2S in *water ice ...*

Sulfur implantation in CO and CO_2 ices

X. Y. Lv[1,2*], P. Boduch[2], J. J. Ding[2], A. Domaracka[2], T. Langlinay[2], M. E. Palumbo[3], H. Rothard[2] and G. Strazzulla[3†]
[1] *School of Nuclear Science and Technology, Lanzhou University, Lanzhou 730000, China*
[2] *Centre de Recherche sur les Ions, les Matériaux et la Photonique (CEA/CNRS/ENSICAEN/UCBN), CIMAP-CIRIL-GANIL, Boulevard Henri Becquerel, BP 5133, F-14070 Caen Cedex 05, France*
[3] *INAF–Osservatorio Astrofisico di Catania, Catania, Italy*

Monthly Notices of the Royal Astronomical Society MNRAS 438 (2014) 922

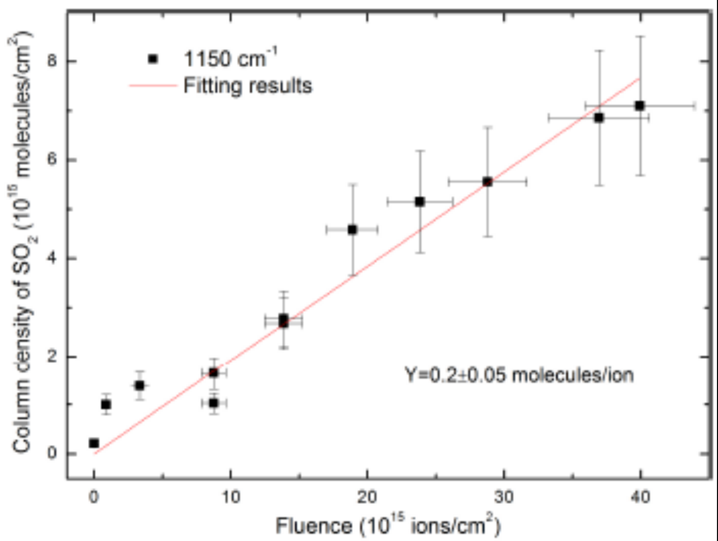


Figure 4. Column density of SO_2 produced after 176-keV S^{11+} implantation in CO ice, as a function of ion fluence. The error bar is about 10 per cent for fluence and 20 per cent for column density.

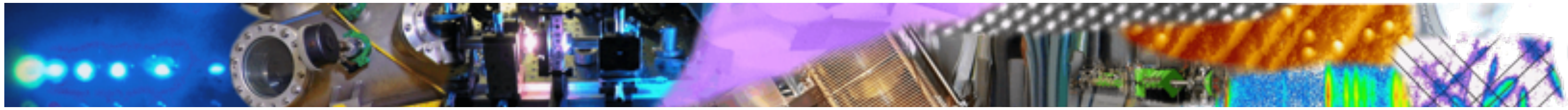
S implantation in CO and CO_2:

Y(**SO_2**) = 0.20 ± 0.05 molec./ion at 176 keV in **CO**
Y(**SO_2**) = 0.38 ± 0.02 molec./ion at 90 keV in **CO_2**

and ... **CS_2** produced in **CO_2** and **OCS** in **CO**

Europa: time to produce observed amount of **SO_2** depends strongly on CO_2 concentration:
200 years ... up to 20000 years

Star forming regions: strong flux of stellar wind from young stars (T-Tauri phase) interacts with CO_2 rich dust, later incorporated in comets



E

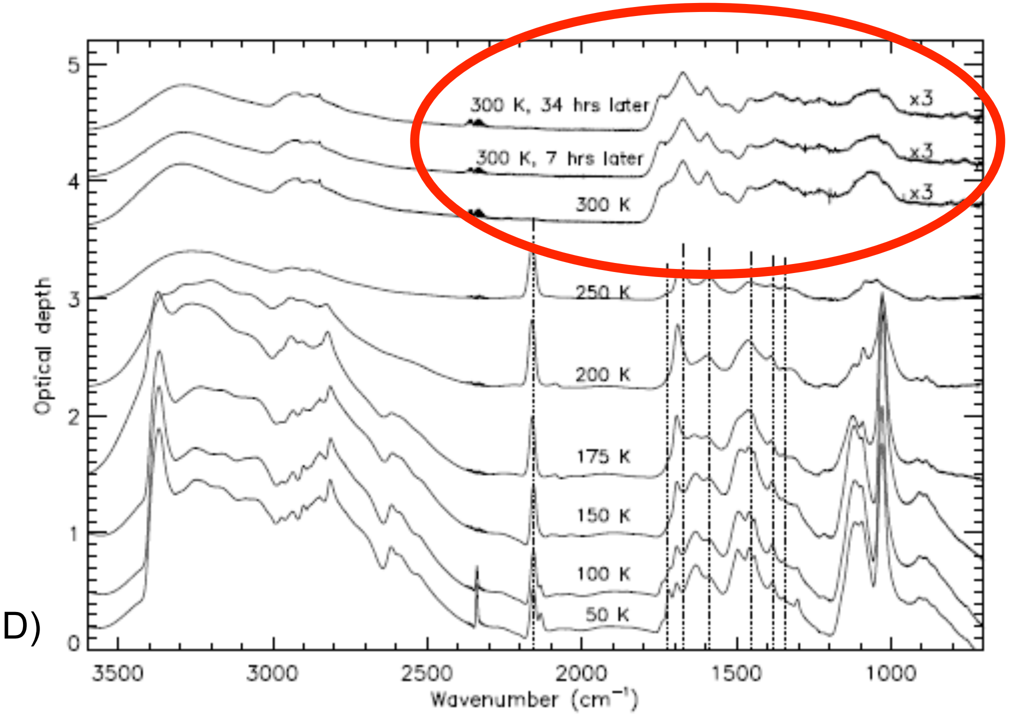
Formation of prebiotic molecules

Radiolysis: formation of prebiotic molecules ?

G. M. Muňoz Caro, E. Dartois,
P. Boduch, H. Rothard,
A. Domaracka, A. Jiménez-Escobar
Comparison of UV and high-energy ion irradiationof methanol:ammonia ice,
Astron. & Astrophys. 566 (2014) A93

NH_3:CH_3OH ice

CASIMIR@GANIL: Zn (SME), Ne (IRRSUD)



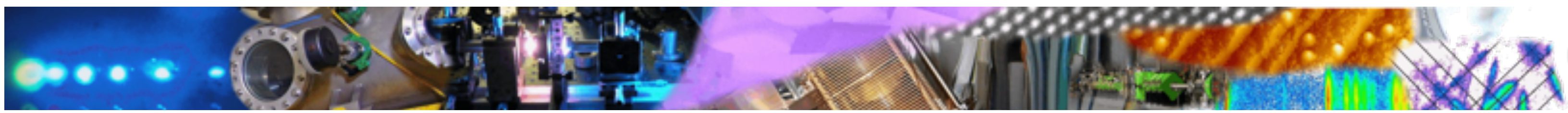
New bands attributed to irradiation products

position[a] (cm^{-1})	Assignment	vibration mode
2340	CO_2	CO str.
2160	OCN^-	CN str.
2138	CO	CO str.
1740	C=O ester/aldehyde	CO str.
1720	H_2CO	CO str.
1694	$HCONH_2$?	CO str.
1587	COO^- in carb. ac. salts[b,c]	COO^- asym. str.
1498	H_2CO	CH_2 scis.
1385	CH_3 groups	CH_3 sym. def.
1347	COO^- in carb. ac. salts[b,c]	COO^- sym. str.
1303	CH_4	def.

at 300K:
stable organic
Residues!

UV vs. heavy ions:
same products

SPIRIT



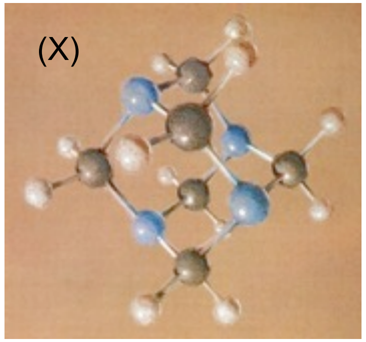
CiMap

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	$NO_3^\dagger$
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)



hexamethylene-tetramine HMT

Donia Baklouti, R. Brunetto, E. Dartois, IAS (Institut d'Astrophysique Spatiale, Orsay), et al.

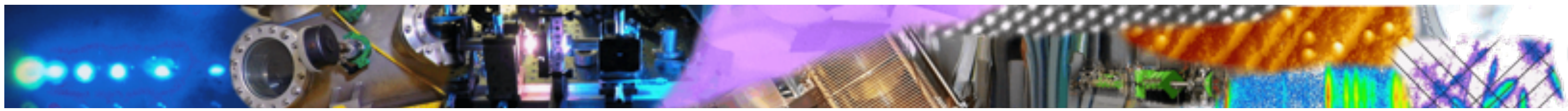
Analysis of the Residues by Chromatography

H_2O- NH_3-CH_3OH ice

(experiment 2014)

Williamary Portugal, Sergio Pilling, Philippe Boduch, Hermann Rothard, Diana Andrade
Radiolysis of Amino Acids by Heavy and Energetic Cosmic Ray Analogs in Simulated Space Environments: α-Glycine Zwitterion Form
Monthly Notices of the Royal Astronomical Society 441 (2014) 3209–3225

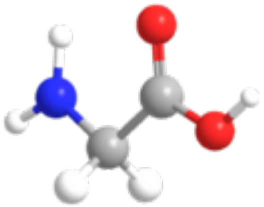
(X) http://osulibrary.oregonstate.edu/specialcollections/coll/pauling/bond/index.html

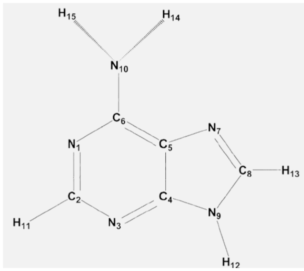


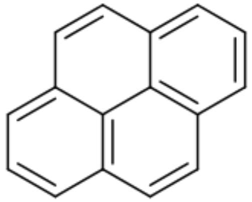
Radiation resistance of organic molecules

⇒ **Irradiation of ices containing complexe molecules**

e.g. glycine, adenine, PAH (Polycyclic aromatic hydrocarbons)







other materials:
carbonaceous, silicates
chemistry at interfaces?

complex radiation field in space: synergy effects?

Emergence of life: Present + future Space Missions

Origin of organic matter in ...

Comets	Rosetta + Philae ESA
Micrometeorites e.g. UCAMMs: (Ultracarboneauceous Antarctic Micrometeroties)	
Asteroids	Hayabusa JAXA
Transneptunian Objects (e.g. Pluto + Charon) + Kuiperbelt Objects	New Horizons NASA
Jovian Moons	JUICE ESA Cosmic Vision launch 2022 (Jupiter Icy Moons Explorer)


aribe
Ciril
CEA - CNRS - ENSICAEN
GANIL
CEA/DSM - CNRS/IN2P3
Jingjie, Xueyang, Gianni, Thomas,
Hermann, Philippe, Stéphane
CiMap
Vinicius, Ana, Sergio
Hussein+Enio
Icy bodies on earth at 270 K:
hydrometeors (winter in Caen)
Eduardo
Emmanuel
Alicja