

Scientific activities in ARIBE the low-energy ion beam facility at GANIL

Patrick Rousseau



- 1 Presentation
- 2 Some statistics
- 3 Some scientific results
 - Instrumental development
 - Dynamics
 - Reactivity
- 4 Perspectives

History

Hall D was initially dedicated to ion source developments.

- 2000 LIMBE, two beamlines for users
- 2005 ARIBE, three additional beamlines
- 2010 GTS ion source is installed
- 2018 GTS upgraded
- 2021 ARIBE updated

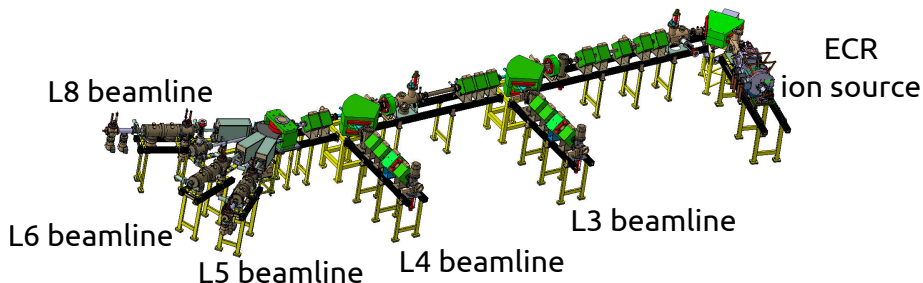


ARIBE: accelerator

ARIBE accelerator is composed by :

- 1 electron cyclotron resonance (ECR) ion source ;
- 5 beamlines where end-stations can be mounted.

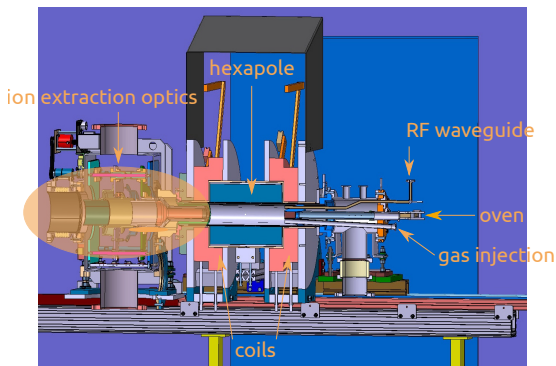
The accelerator was **updated in 2021** (vacuum and ion optics ctrl/cmd).



ARIBE: GTS ions source

The **GTS ECR ion source** allows to produce a large variety of ions with **different charge state**: He^{2+} , O^{6+} , Ar^{11+} , Xe^{25+} ...

Typical **acceleration voltage**: 10-15 kV

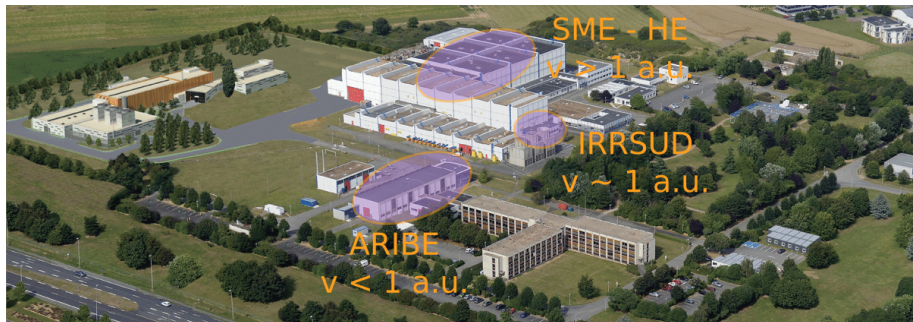


Ion beam map

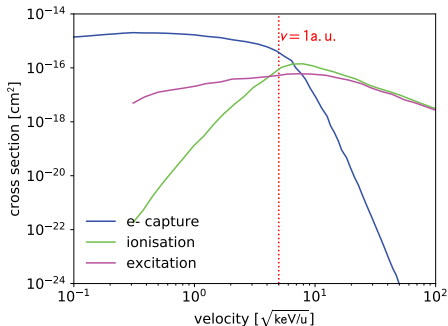
ARIBE is the low-energy ion beam facility of **GANIL**.

It delivers **slow ions**, i.e. with velocity below one atomic unit of velocity, thus slower than electrons in matter.

It complements the “offer” to interdisciplinary physics users. *Civil*



Electronic processes: cross sections



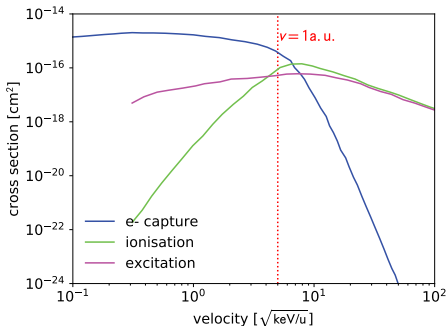
Collisions $\text{H}^+ - \text{H}$

[D. Vernhet et al., *Nucl. Instrum. Meth. B* **107** (1996) 71]

Electronic processes: cross sections

For **low velocity**, i.e. below the Bohr velocity, the **electron capture** dominates.

$$\sigma_{\text{e-capt}} \sim 10^{-15} - 10^{-14} \text{ cm}^2$$



Collisions H⁺ – H

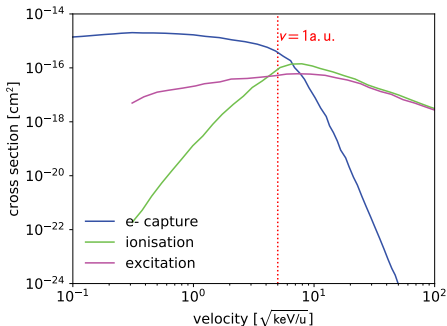
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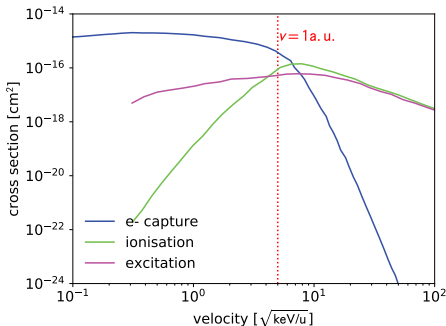
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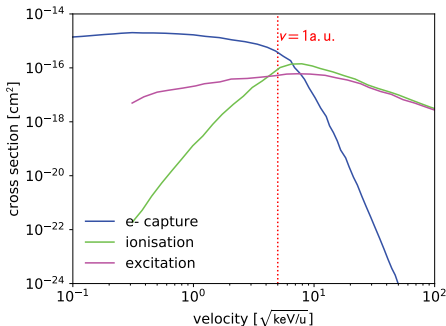
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Excitation occurs all over the whole velocity range.

In the **intermediate regime**, all processes occur with similar cross sections.

- maximum energy deposit
- **Bragg peak**



Collisions H⁺ – H

[D. Vernhet et al., *Nucl. Instrum. Meth. B* **107** (1996) 71]

Electron capture

ARIBE delivers low-energy (few keV) ions thus **slow** ($v < 1$ a.u.).

The (resonant) **electron capture** is the dominant process.



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Classical-over-barrier (COB) model

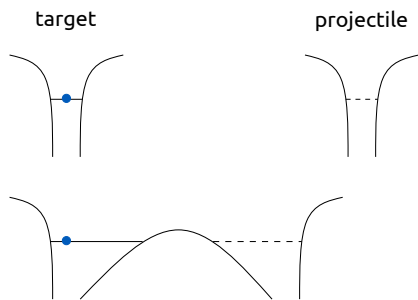


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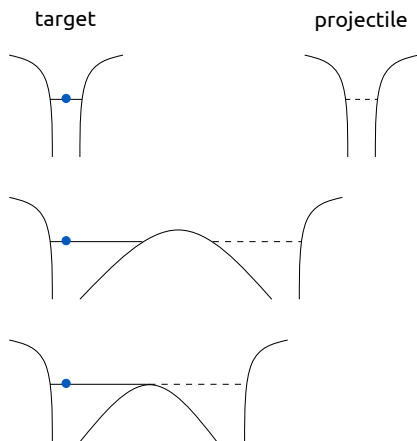


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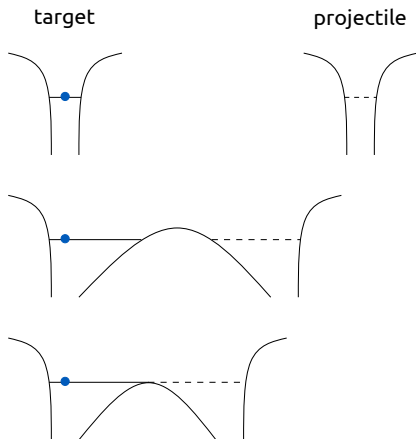
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Classical-over-barrier (COB) model

Stronger barrier lowering with higher projectile charge state

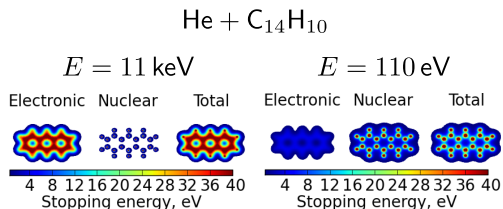
With multiply charged ions, (multiple) capture at **large distance** → **“cold” molecular ions**

Large yield of multiply charged molecular cations



Energy loss: stopping power

Due to its mass, an atomic projectile can interact with both electrons and nuclei:

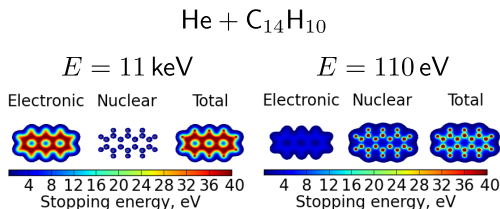


[Tao Chen, PhD thesis, Stockholm University]

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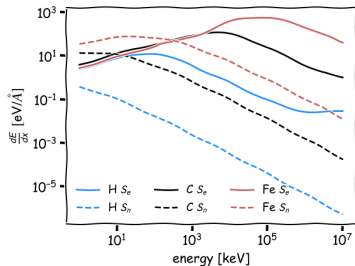
- the **nuclear energy loss** is due to elastic binary nucleus-nucleus collisions



[Tao Chen, PhD thesis, Stockholm University]

Energy loss in anthracene C₁₄H₁₀

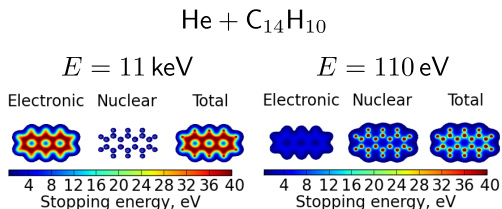
[calculated by SRIM <http://www.srim.org/>]



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Due to its mass, an atomic projectile can interact with both electrons and nuclei:

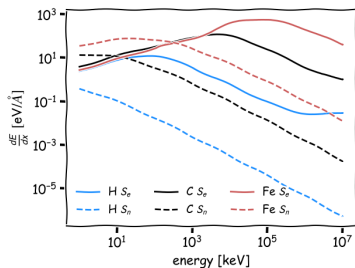
- the **nuclear energy loss** is due to elastic binary nucleus-nucleus collisions
- the **electronic energy loss** is due to the friction of the projectile on the electronic cloud



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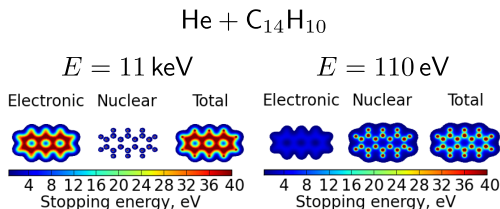
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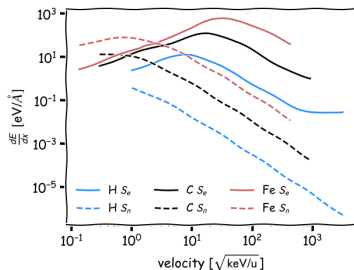
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Take home message #1

Ion interaction leads to:

1 electron removal

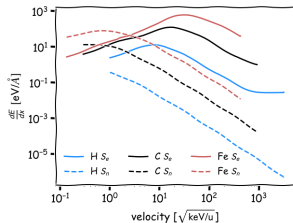
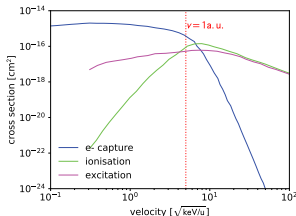
- by electron capture at low velocity
- by ionisation at higher velocity

2 energy deposit

- by nuclear energy loss at low velocity
- by electronic energy loss at higher energy

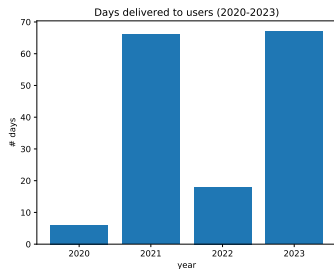
With **ARIBE beams**, electron capture dominates and nuclear energy loss is not negligible.

ARIBE offers a **flexibility** in the choice of **projectile nature and charge state**.

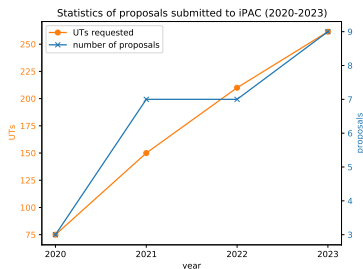
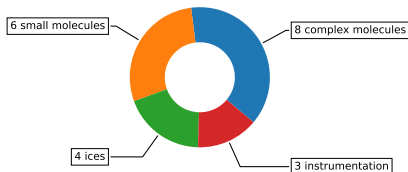


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2020-2023 statistics



Publications using ARIBE beams (2020-2023)



One PhD per year at CIMAP
plus PhD students of users.

Users come from:

Caen CIMAP, GANIL

France Paris, Marseille, Grenoble...

Europe Italy, Sweden, Czech Republic...

World India, Brazil, USA...

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Instrumentation developed for FISIC project

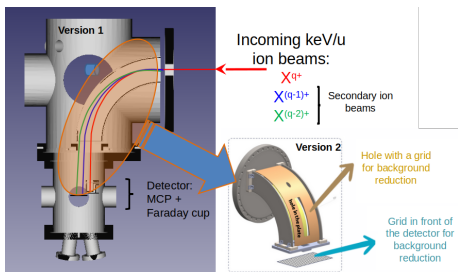
FISIC: Slow ion - fast ion collisions

→ measurements of **cross sections in the intermediate region**

Test of instruments at ARIBE in 2021 and 2023.

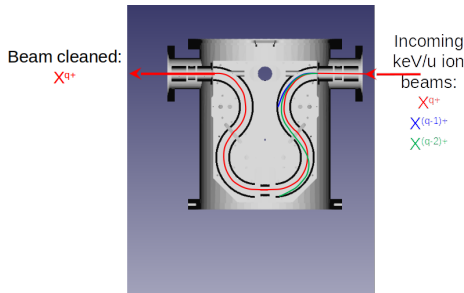
Ion spectrometer

[M. Jolly et al., *Atoms* **10** (2022) 146]



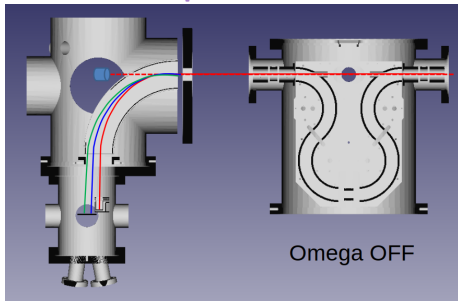
Charge state purifier

[D. Schury et al., *Rev. Sci. Instrum.* **90** (2019) 083306]

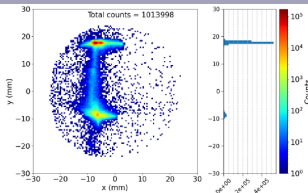
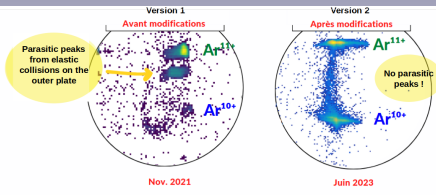
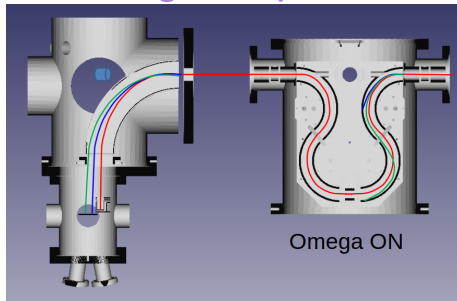


Instrumentation developed for FISIC project (2)

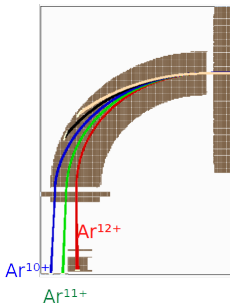
Ion spectrometer



Charge state purifier



Instrumentation developed for FISIC project (3)



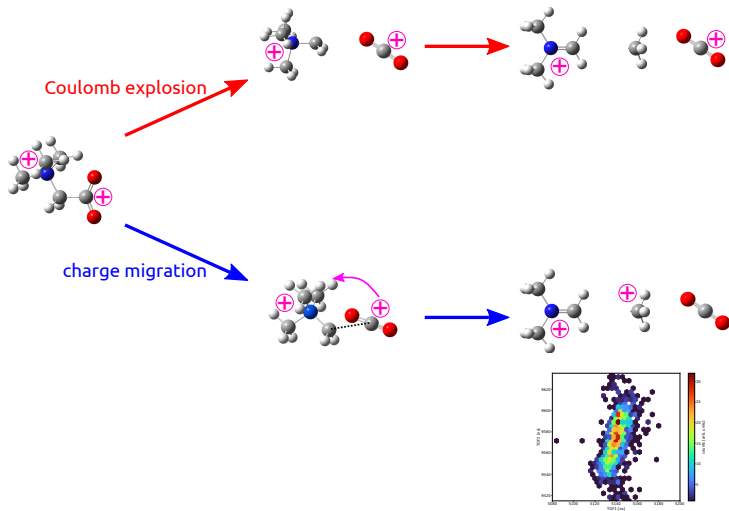
From the recorded counts on the MCP (Ar^{11+} , Ar^{10+}), the intensity of the main beam (Ar^{12+}) on the Faraday cup, and a set of coupled differential equations, it is possible to extract **capture cross sections** from N_2 .

process	calculated CS	extracted CS
$Ar^{12+} \rightarrow Ar^{11+}$	$1.33 \cdot 10^{-14} \text{ cm}^2$	$(1.43 \pm 0.74) \cdot 10^{-14} \text{ cm}^2$
$Ar^{12+} \rightarrow Ar^{10+}$	$2.88 \cdot 10^{-15} \text{ cm}^2$	$(1.78 \pm 0.95) \cdot 10^{-15} \text{ cm}^2$

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Charge migration vs. Coulomb explosion

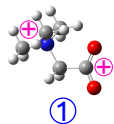
Beside the expected **“Coulomb explosion”** we observe **charge migration**.



Scenario of charge migration in betaine dication

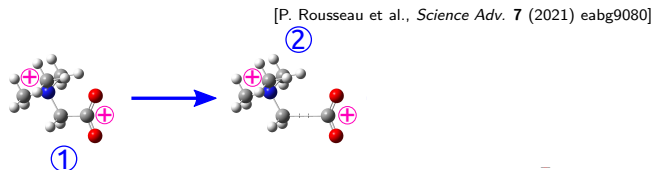
- ① **two charges localised at both ends** due to electron captures

[P. Rousseau et al., *Science Adv.* **7** (2021) eabg9080]



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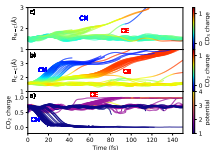
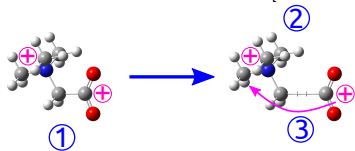
- 1 **two charges localised at both ends** due to electron captures
- 2 **charge repulsion and elongation** of the C – C_α bond



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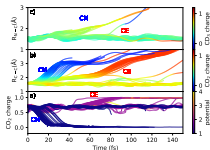
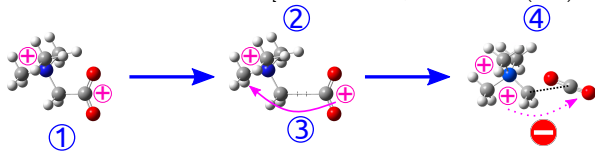
[P. Rousseau et al., *Science Adv.* 7 (2021) eabg9080]



Scenario of charge migration in betaine dication

- ① **two charges localised at both ends** due to electron captures
- ② **charge repulsion and elongation** of the C – C_α bond
- ③ **charge migration**
- ④ formation of a **transient species cutting the way back** for charge

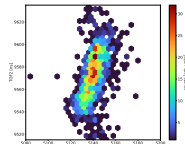
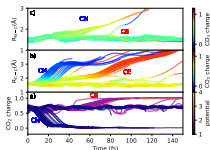
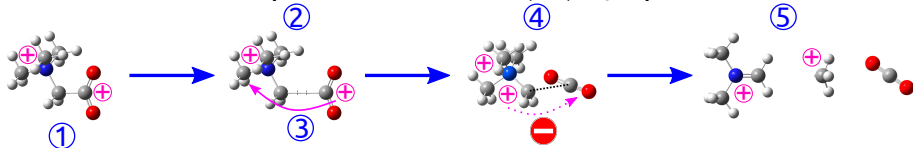
[P. Rousseau et al., *Science Adv.* 7 (2021) eabg9080]



Scenario of charge migration in betaine dication

- 1 **two charges localised at both ends** due to electron captures
- 2 **charge repulsion and elongation** of the C – C_α bond
- 3 **charge migration**
- 4 formation of a **transient species cutting the way back** for charge
- 5 emission of **two charged fragments** in the **same direction**

[P. Rousseau et al., *Science Adv.* 7 (2021) eabg9080]



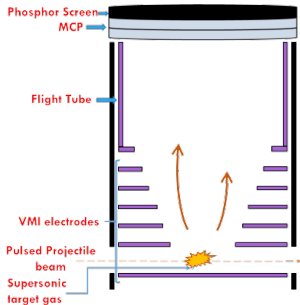
Ion-induced molecular fragmentation: case of N_2

Measuring **fragment ion KE and emission angle** with a **Velocity Map Imaging spectrometer**.

→ information on collision processes and fragmentation dynamics

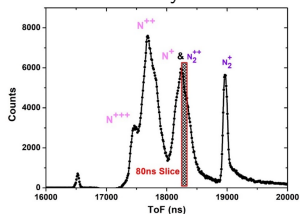
IMAGERI VMI spectrometer

[N. Sens et al., *Rev. Sci. Instrum.* **93** (2022) 085103]



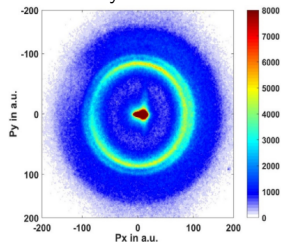
Time-of-Flight mode

→ identify ions



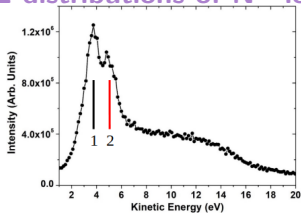
Imaging mode

→ velocity of selected ions

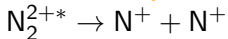


Ion-induced molecular fragmentation: case of N_2 (2)

KE distributions of N^+ ions

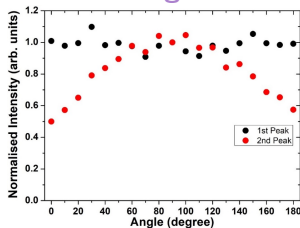


Coulomb explosion



- To understand the **orientation dependence** of N_2^{2+*} dissociation
- collision processes (electron capture, transfer ionisation etc.) depend on **projectile charge state and velocity**
 - iPAC2023 : 12keV- He^+ + N_2 (ARIBE); 35MeV- Ar^{10+} + N_2 (IRRSUD)

Emission angle vs. KE



Second peak shows **anisotropy** distribution peaking at 90°

Take home message #2

Collisions with slow multiply charged ions allows to produce **multiply charged molecular ions**.

The problem of the distributions of initial states can be overcome using **coincidence measurements**.

“**Coulomb explosion**” of the molecular systems can give information on the **dynamics timing** even without pump-probe scheme.

→ **Nuclear dynamics** observed (e.g. atom migration)

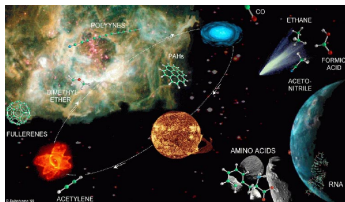
→ **Electronic dynamics** observed (e.g. ICD, charge migration)

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Molecules in space

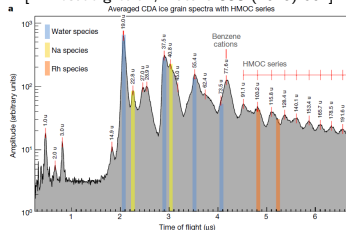
We are living in a **molecular universe**.

Both **neutral and ionic species** have been identified (+200 species).
Complex organic molecules observed in meteorites, on Saturn moons.



Enceladus dust particle mass spectra

[F. Postberg et al., *Nature* **558** (2018) 564]

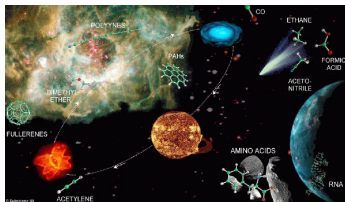


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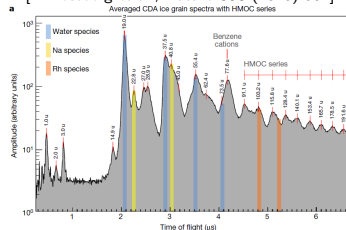
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Molecules are **exposed to ionising particles** (photons, electrons, ions).
What is the role of **energetic processing by radiation** on growth processes?



Enceladus dust particle mass spectra

[F. Postberg et al., *Nature* **558** (2018) 564]



Bottom-up, Top-down, What else?

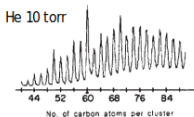
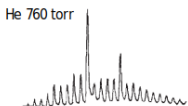
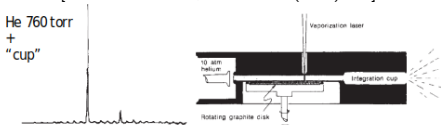
Two approaches are usually considered to produce complex molecules.

Starting from small units to grow

→ **“bottom-up”**

Laser vaporisation of graphite

[H. W. Kroto et al., *Nature* **318** (1985) 162]

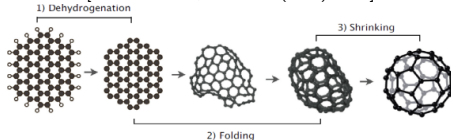


Processing large particles of matter

→ **“top-down”**

Processing of large PAH

[O. Berné et al., *A&A* **577** (2015) A133]



Bottom-up, Top-down, What else?

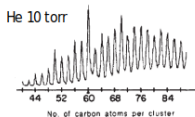
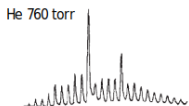
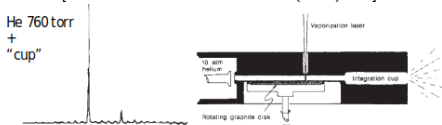
Two approaches are usually considered to produce complex molecules.

Starting from small units to grow

→ **“bottom-up”**

Laser vaporisation of graphite

[H. W. Kroto et al., *Nature* **318** (1985) 162]

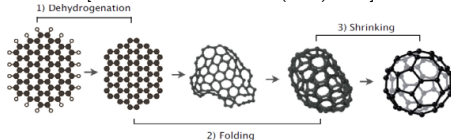


Processing large particles of matter

→ **“top-down”**

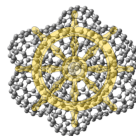
Processing of large PAH

[O. Berné et al., *A&A* **577** (2015) A133]



“middle way”

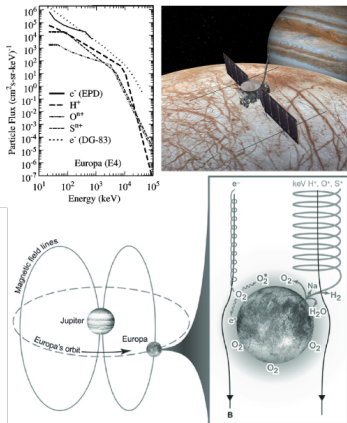
→ reactivity inside molecular clusters



Implantation of sulfur ions in ices

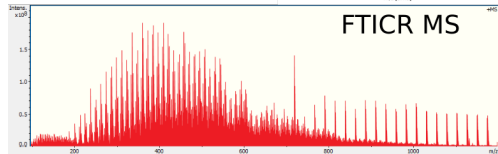
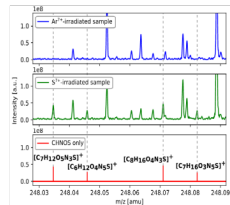
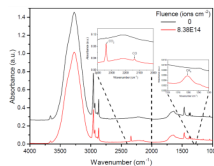
Europa is impacted by low energy ions from Jupiter magnetosphere.

Production of **organosulfur compounds** by implantation of **S^{q+} ions**?



Irradiation of $H_2O : C_3H_8$ ice
by 105 keV S^{7+} ions

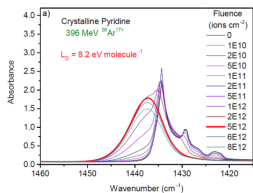
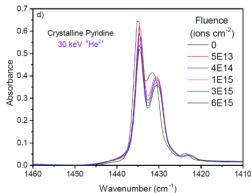
FTIR spectroscopy



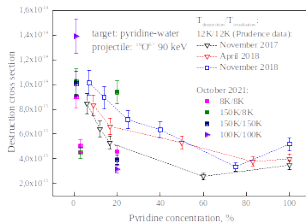
Irradiation of complex organic molecules

Amorphisation and destruction of COM ices on planet moons

Amorphisation of pyridine ices



Destruction of pyridine ices



Preliminary results show:

- amorphisation by SHI (Ar@SME)
- no amorphisation by low-energy He²⁺

→ looking for the **effect of heavier** (O, S) low-energy ions

Higher destruction yield in **mixed ices** water:pyridine
→ need to consider mixed ices in models

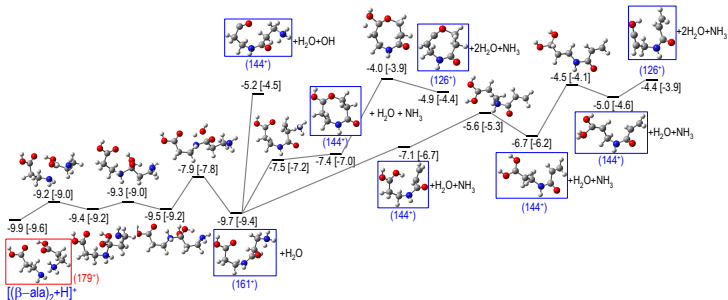
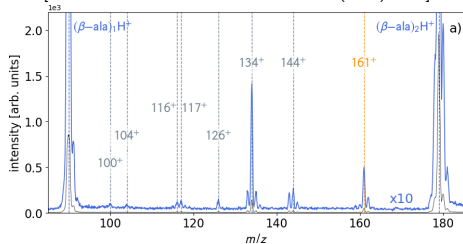
Formation of peptide bonds

Successive losses of 18 mass units
from the β -alanine clusters
→ peptide bond formation?

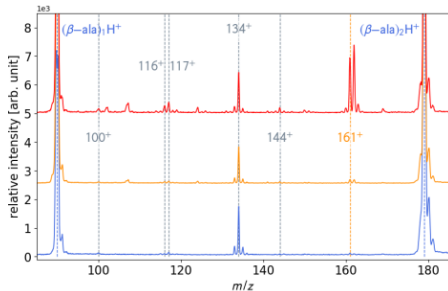
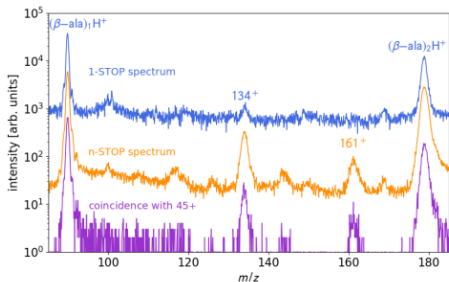
Similar patterns observed
→ same mechanisms?

Potential energy surface
exploration

30 keV He^{2+} collisions with $[\beta - \text{ala}]_k$
[P. Rousseau et al., *Nature Commun.* 11 (2020) 3818]



Formation of peptide bonds (2)



More peptide bond formation:

- for **multiply charged clusters** → role of charge, energy transfer
- for **larger initial clusters** → excess energy is dissipated

Peptide bond formation also observed in glycine clusters (July 2023)

Looking for **chiral effect** in peptide bond formation (March 2024)

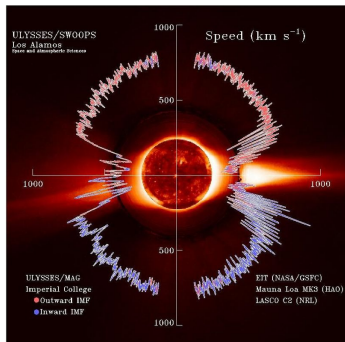
Take home message #3

Ion collision induced **reactivity** observed in both condensed (ices) and gas phase (clusters)

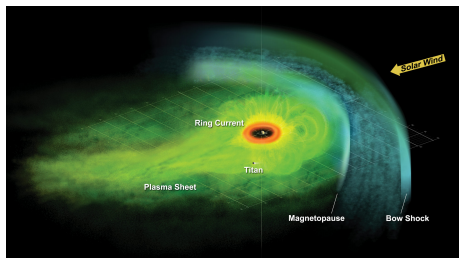
→ **formation of complex molecules**

→ **role of low energy ions** in space molecular inventory wealth

Solar wind



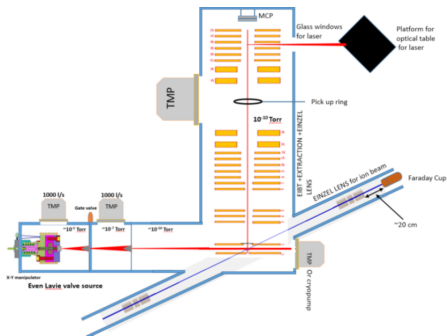
Giant planet magnetosphere



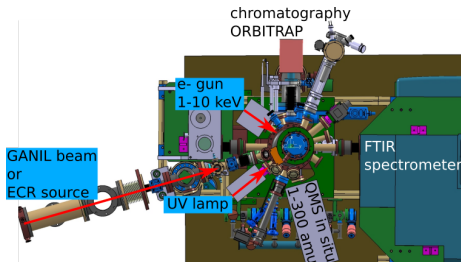
- 1 Presentation
- 2 Some statistics
- 3 Some scientific results
 - Instrumental development
 - Dynamics
 - Reactivity
- 4 Perspectives

New instruments for reactivity studies

FRAPA



MIRRPLA



ANR and FEDER

Know-how acquired could be used to
**develop molecular ion storage
device** for users.

PEPR Origins (France 2030)

Unique device to study the
**irradiation of astrophysical ices
analogues**

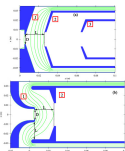
ARIBE: a solar wind simulator

low energy extraction

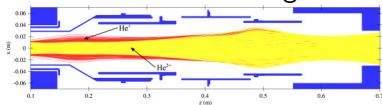
(collaboration IAP Nizhny Novgorod)

[S. S. Vybin et al., *Nucl. Instrum. Meth. A* **1061** (2024) 169109]

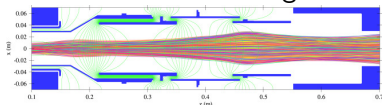
new electrode design



He^{q+} extraction voltage 2 kV



Xe^{q+} extraction voltage 1 kV



“metallic” ions

There is a need to produce new ion beams such as:

- Mg
- Si
- Fe

→ to install the MIVOC method on the GTS source

Alternative solution to produce sulfur ions S^{q+} instead of using SF_6

ARIBE: agreement renewal

To fulfill the next agreement of French Nuclear Safety Agency in **2026**, it is **compulsory** to:

- design a new **safety interlock chain** with modern components
- build a new **shed around the ion source** to have a public area in the whole experimental hall
 - campaigns to measure the radiation emitted by the source (Nov/Dec 2023, April 2024)
 - design of the new shed for summer 2024
 - delivery of the new shed for summer 2025

Thanks to all staff

GANIL

Ciril

