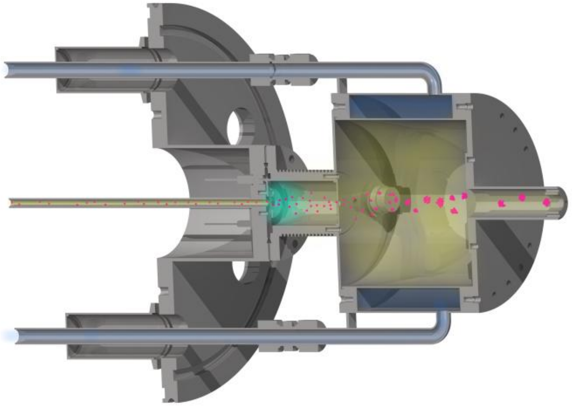
Source à Agrégation Gazeuse pour la Production d'Agrégats Moléculaires Mixtes Thermalisés

Sébastien Zamith, Jean-Marc L'Hermite
Cluster Team



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Outline

- Introduction / context
- Experimental setup
- Gas aggregation source
- Homogeneous clusters
- Heterogeneous clusters
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Introduction / context

- Ensemble of molecules
- Size 2 – 10^3 molecules
- van der Waals / hydrogen bonds
- Mass selection ➔ charged species
- Fragile ➔ cold environment
- Temperature dependence ➔ thermalization

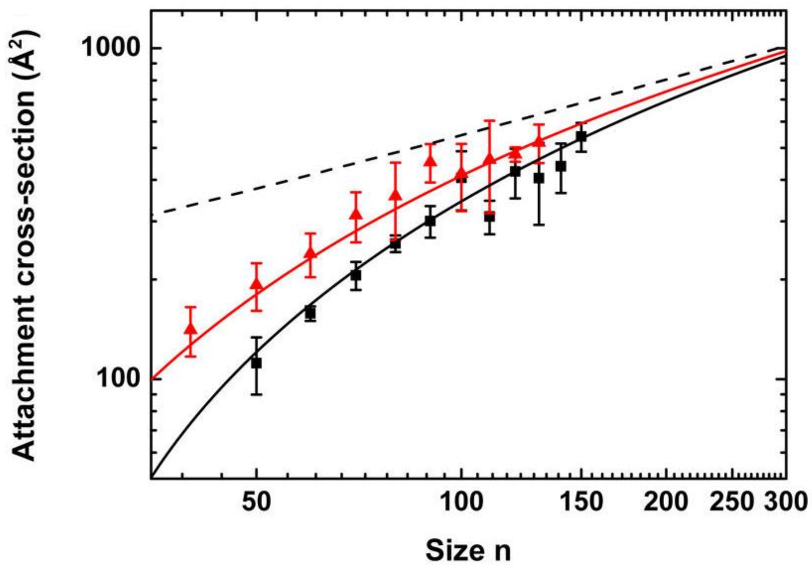
Introduction / context

- Model for microhydration
- Water clusters containing small biomolecules (RNA bases)
- Atmospheric relevant species (H_2SO_4, NH_3)
- Pollutants (PAHs)
- Charged clusters ➔ proton transfer

PAH = polyaromatic hydrocarbons

Introduction / context

Pure alcohols clusters $(CH_4O)_n$, $(C_2H_6O)_n$

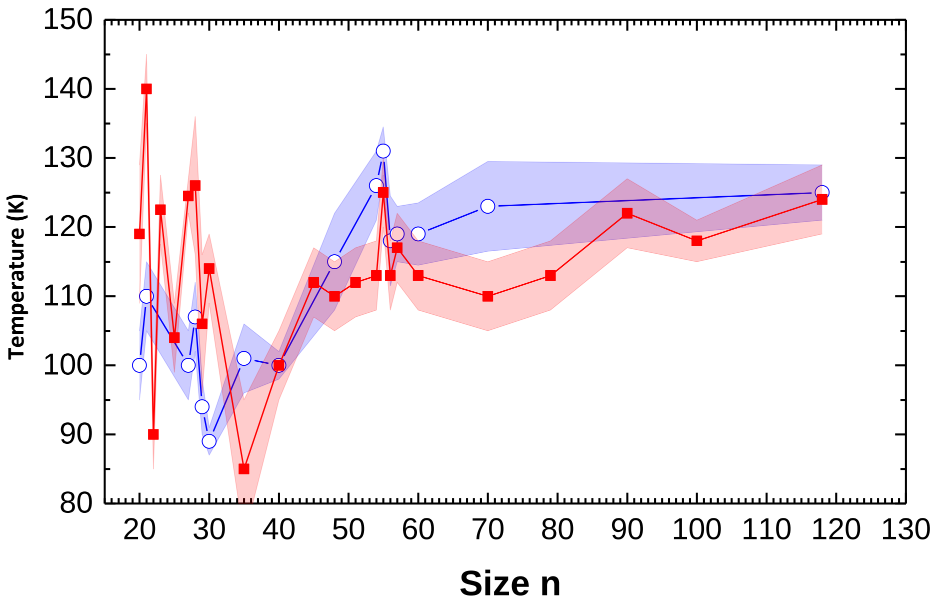


Size dependence of attachment cross section

I. Braud *et al*, J. Phys. Chem. A **119**, 6017 (2015)

Introduction / context

Pure protonated and deprotonated water clusters $(H_2O)_nH^+$, $(H_2O)_{n-1}OH^-$

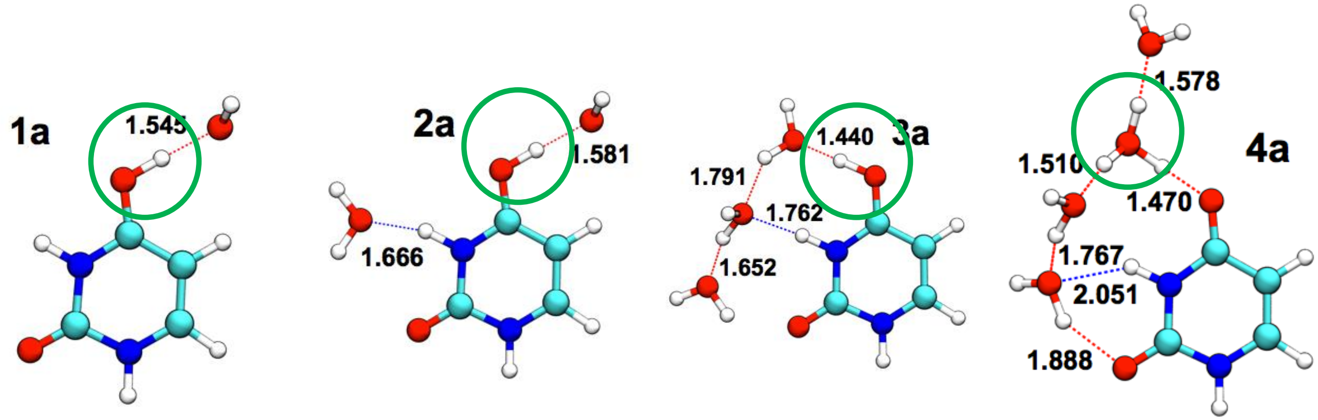


Size dependence of the transition temperature

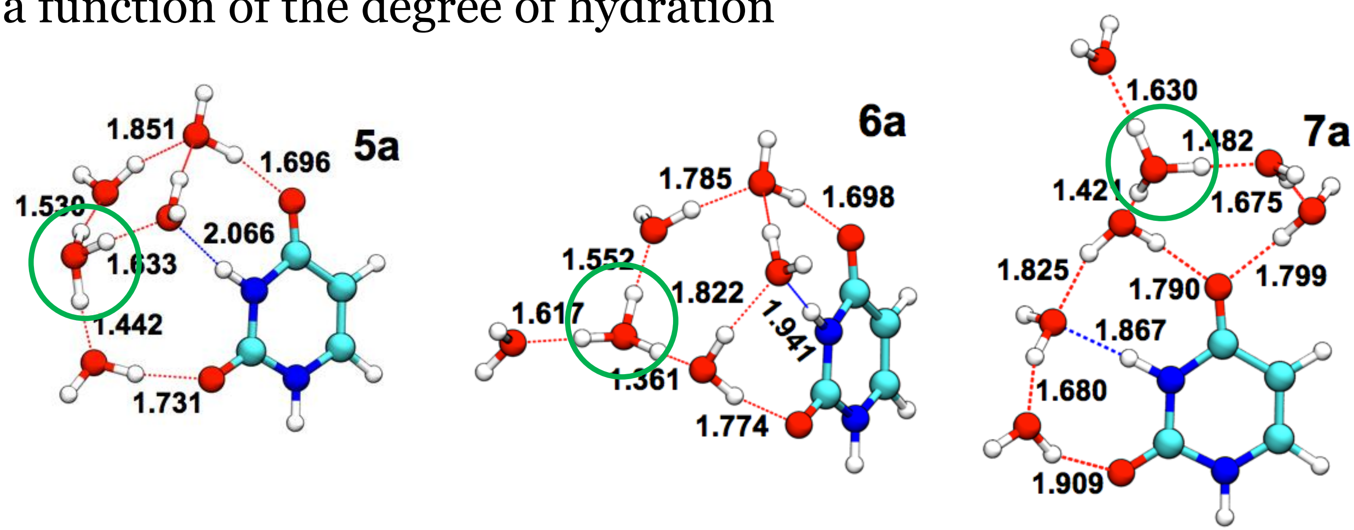
S. Zamith *et al*, J. Chem. Phys. **138**, 034304 (2013)
J. Boulon *et al*, J. Chem. Phys. **140**, 164305 (2014)
K. Korchagina *et al*, Phys. Chem. Chem. Phys. **19**, 27288 (2017)

Introduction / context

Mixed water-uracil clusters $(H_2O)_nUH^+$,



Proton localization as a function of the degree of hydration



I. Braud *et al*, J. Chem. Phys. **150**, 014303 (2019)

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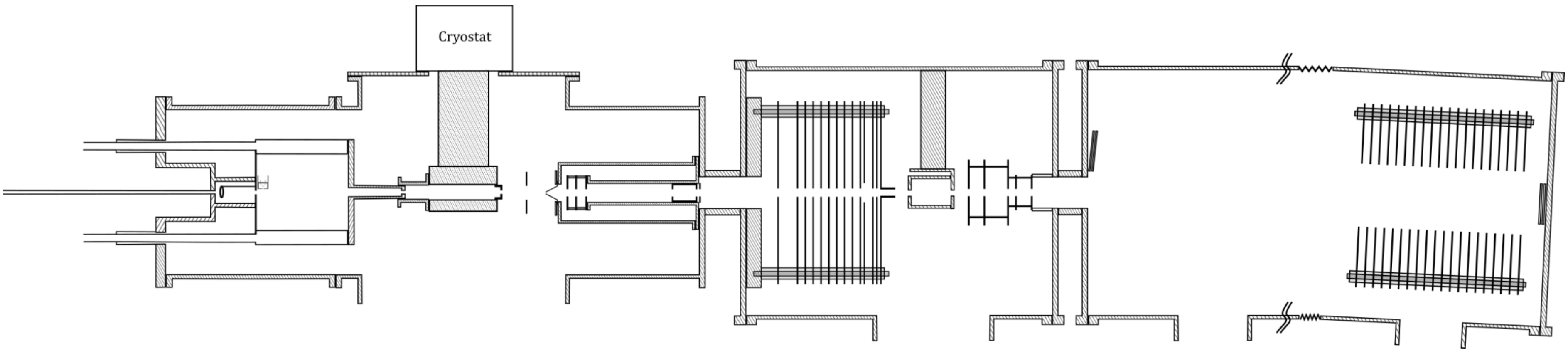
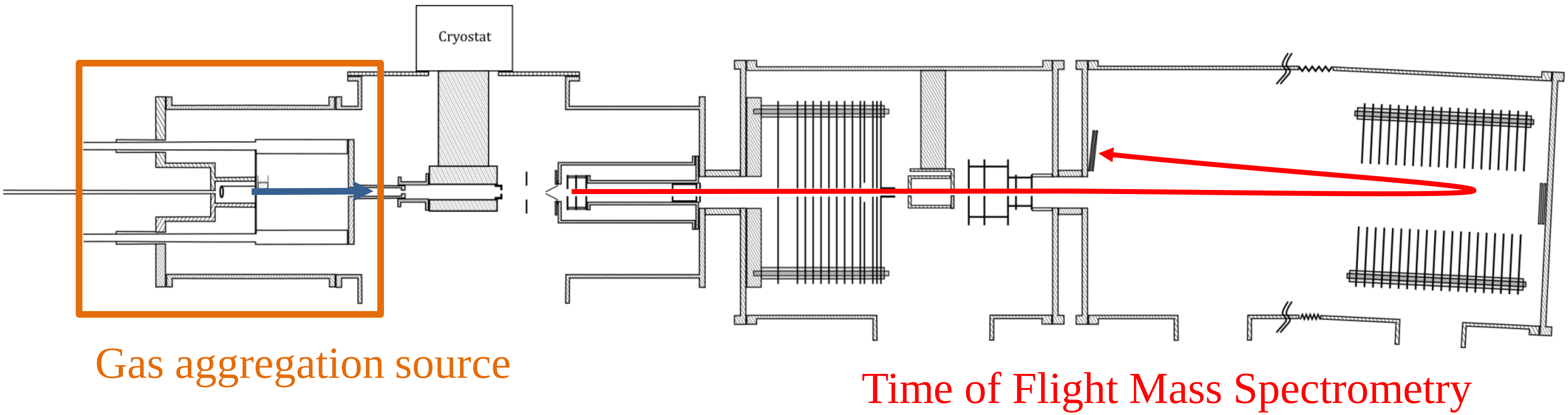
Experimental setup



Experimental setup designed to perform collisions between mass selected clusters and atomic or molecular vapor (attachment cross-section, fragmentation cross-section, nanocalorimetry, spontaneous thermal evaporation, …).

F. Chirot *et al*, Rev. Sci. Instrum. **77**, 063108 (2006)

Experimental setup



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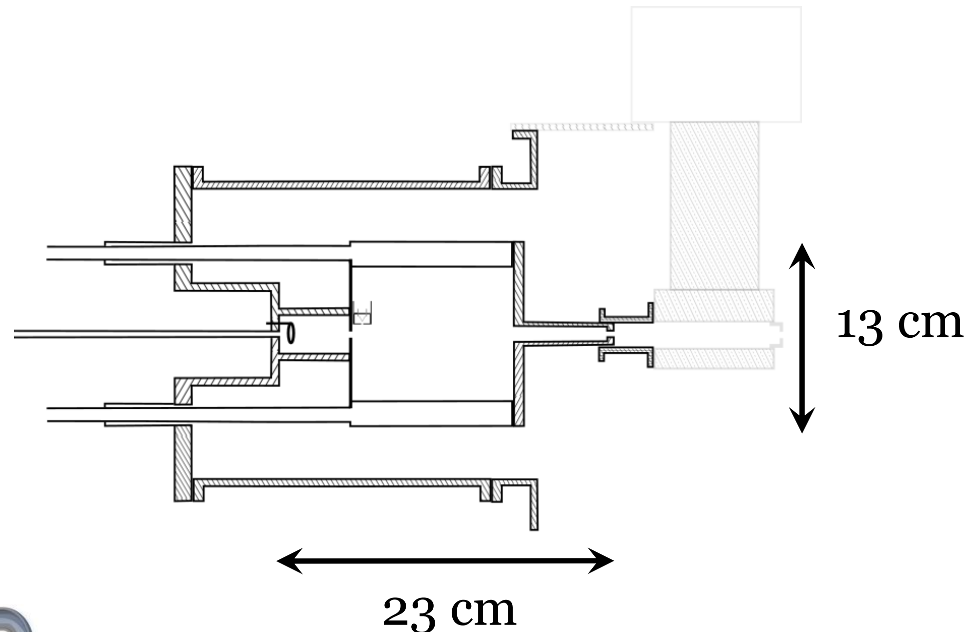
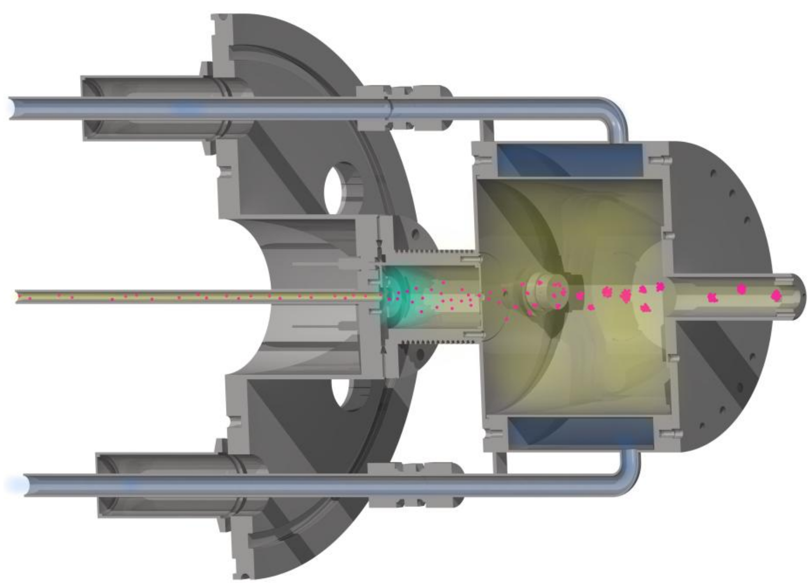
Gas aggregation source

Stainless steel chamber

ISO KF flange DN200

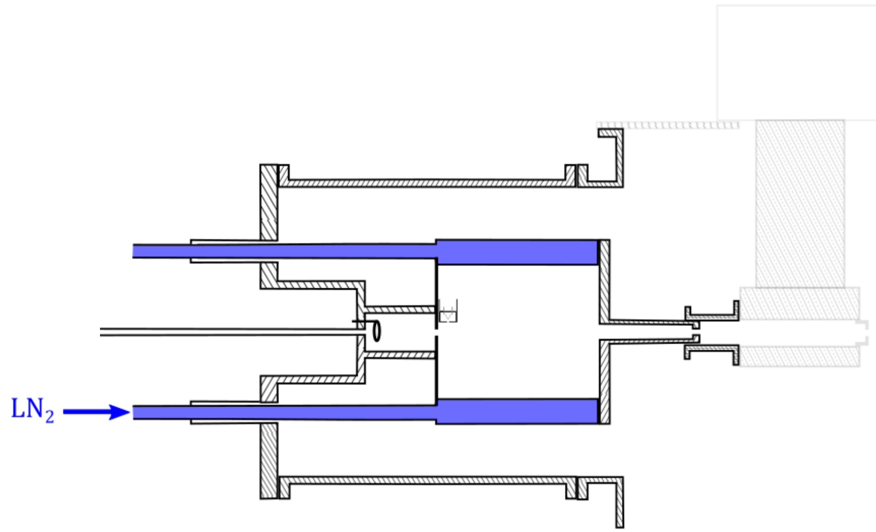




Condensation of a vapor in a cold atmosphere

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

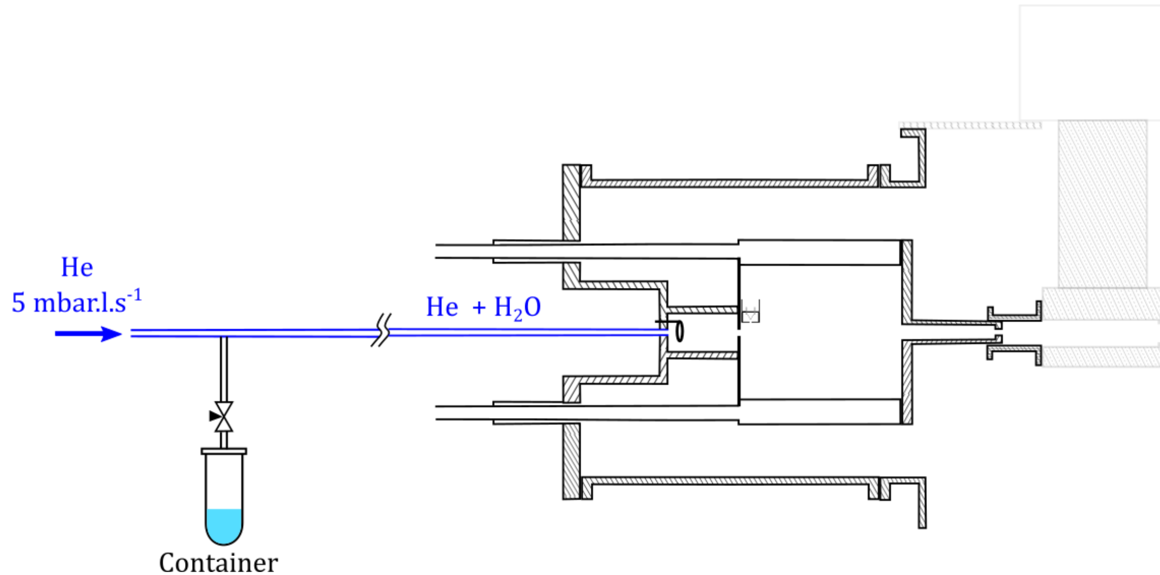
Gas aggregation source



Double wall chamber
→ Liquid nitrogen circulation

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

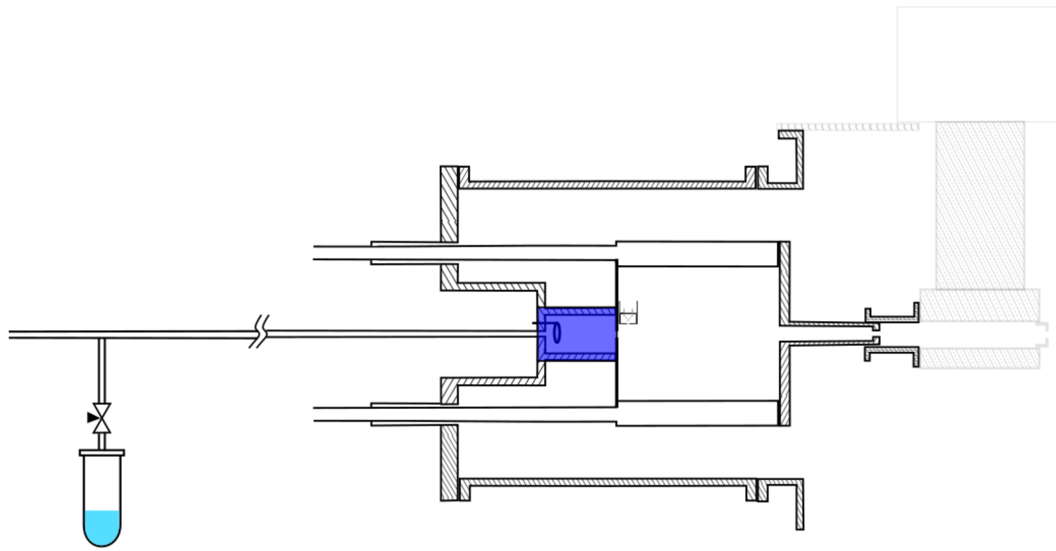
Gas aggregation source



Helium buffer gas injection
→ adjunction of water or other high vapor pressure species

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

Gas aggregation source



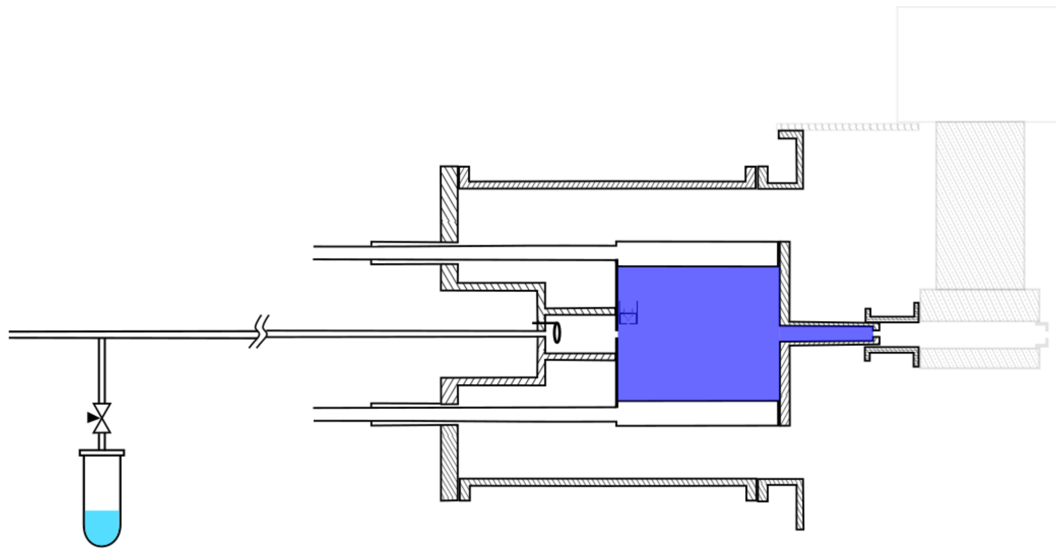
Oven, controlled temperature T = 273 to 425 K

→ low vapor pressure species

→ P ~ 1 mbar

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

Gas aggregation source



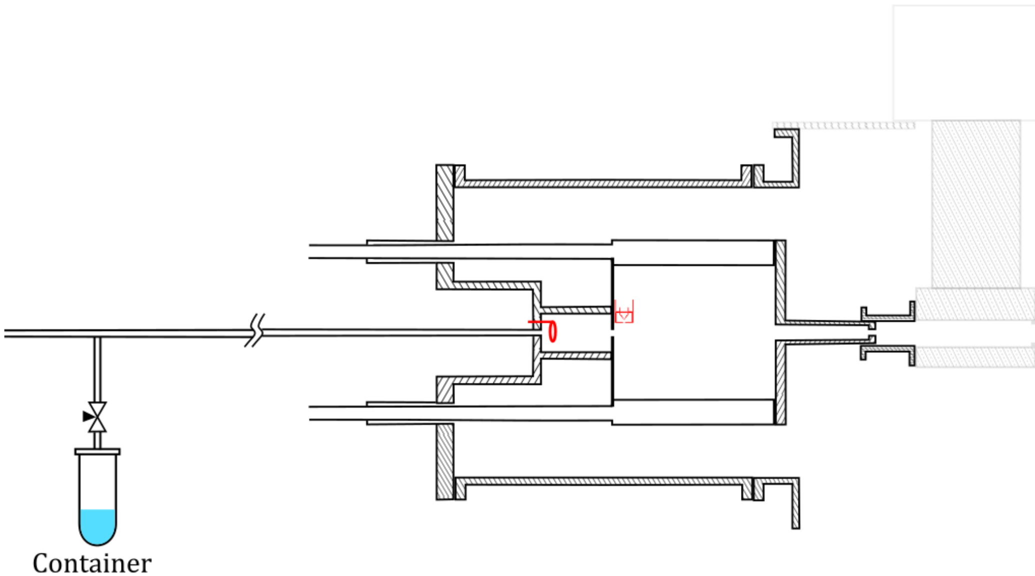
Growth, T = 110 to 90 K

→ Helium flux brings the clusters toward the thermalization chamber

→ P ~ 1 mbar

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

Gas aggregation source

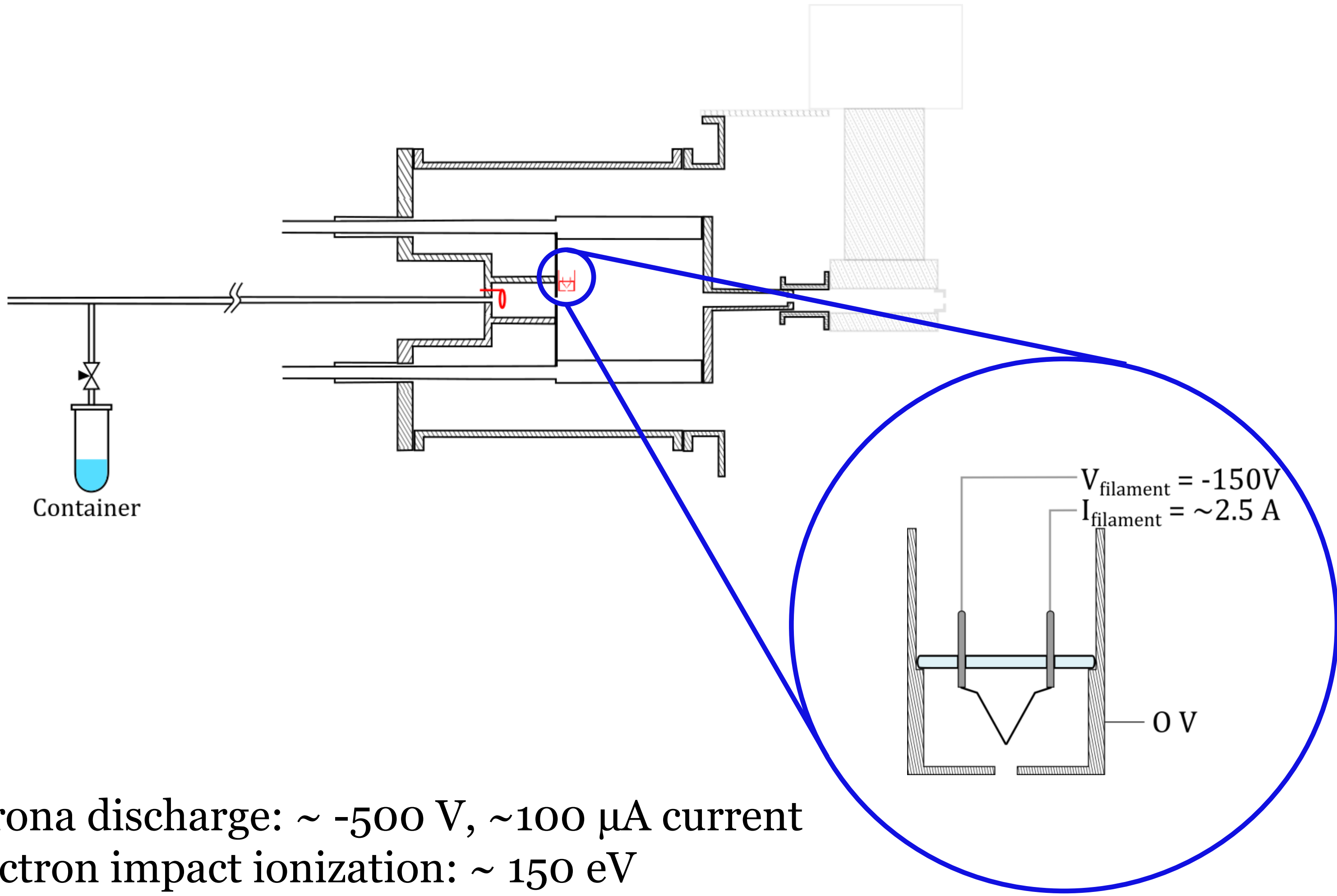


Ionization

→ Corona discharge: ~ -500 V, ~100 μA current
→ Electron impact ionization: ~ 150 eV

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

Gas aggregation source



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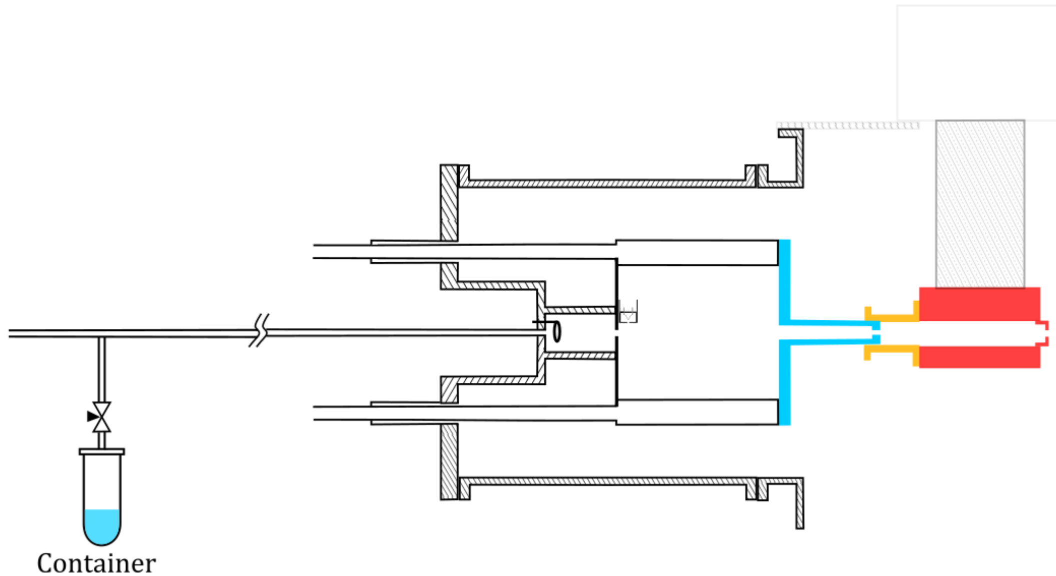


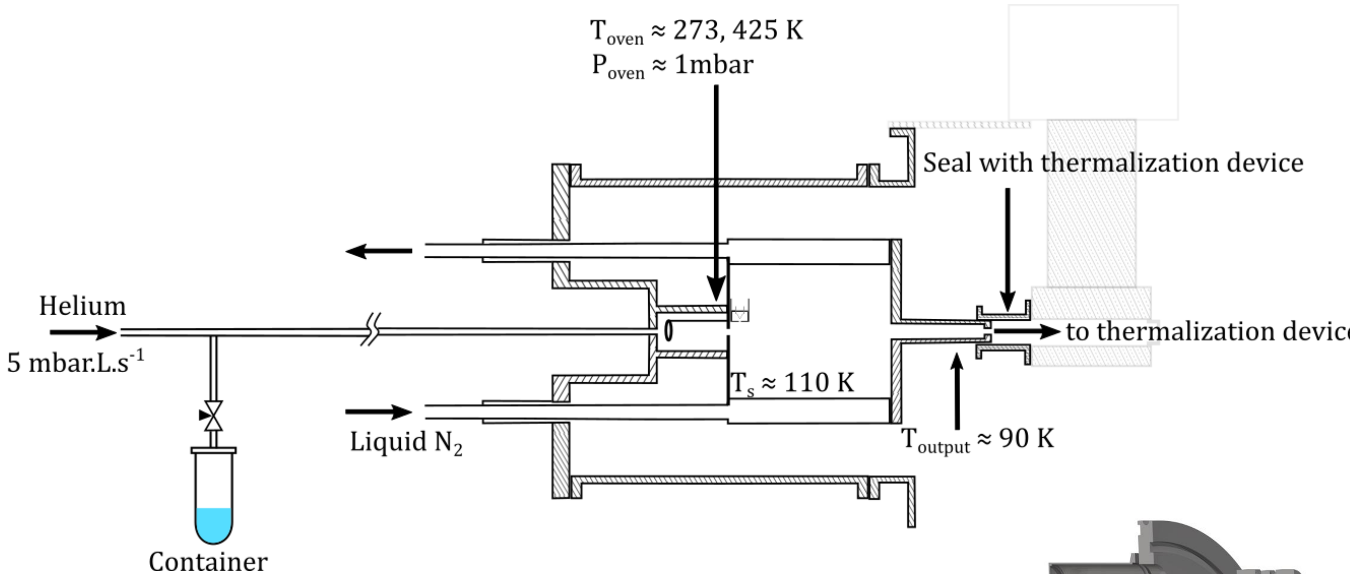
Materials:

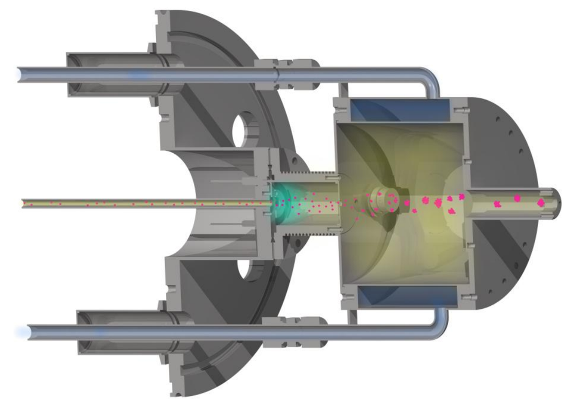
Stainless steel
Aluminium
PEEK
Copper

Indium wire

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

Gas aggregation source





Adjustable parameters:

- needle valve opening
- oven temperature
- helium flux
- source and thermalization chamber exit diameters
- discharge intensity / electron gun current

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)

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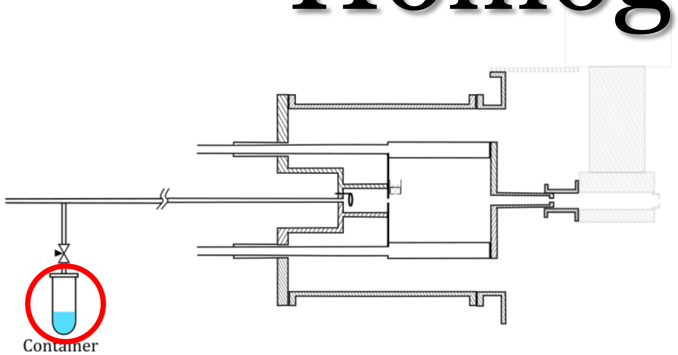
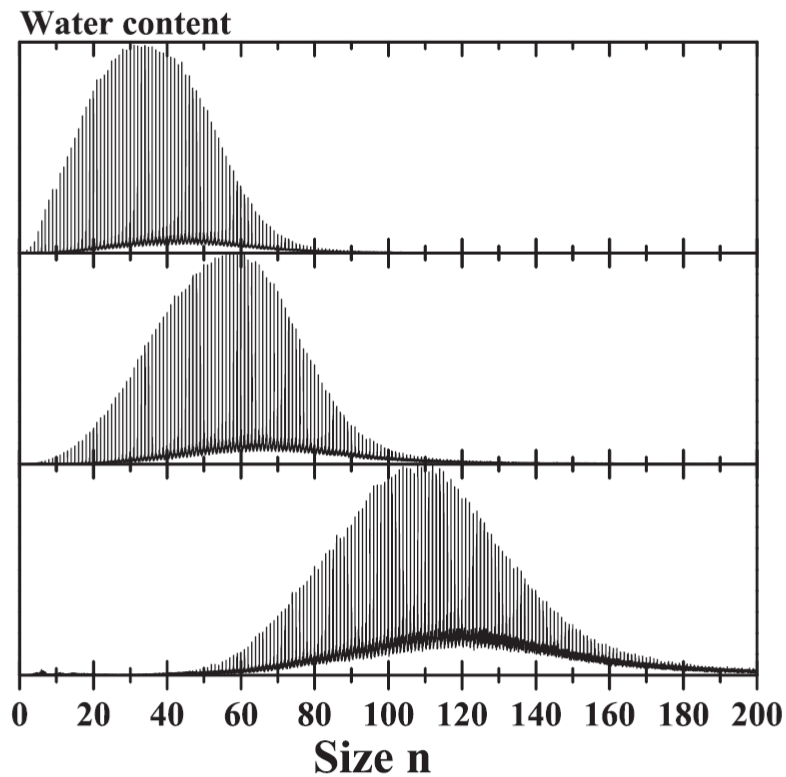
Homogeneous clusters





Influence of water content

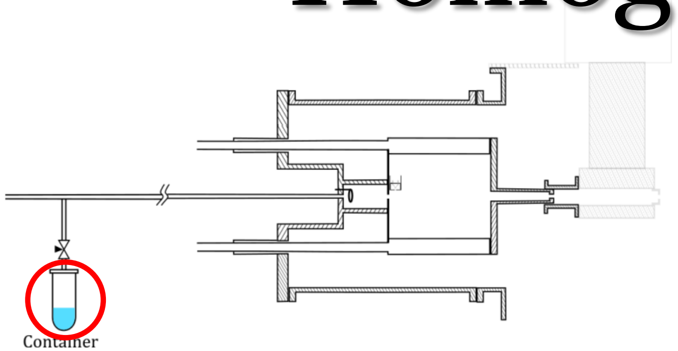
More material available ➔ larger clusters

Discharge ionization

$P(H_2O)$ = 23 mbar @ 293K

~ 1 % water in helium

Homogeneous clusters



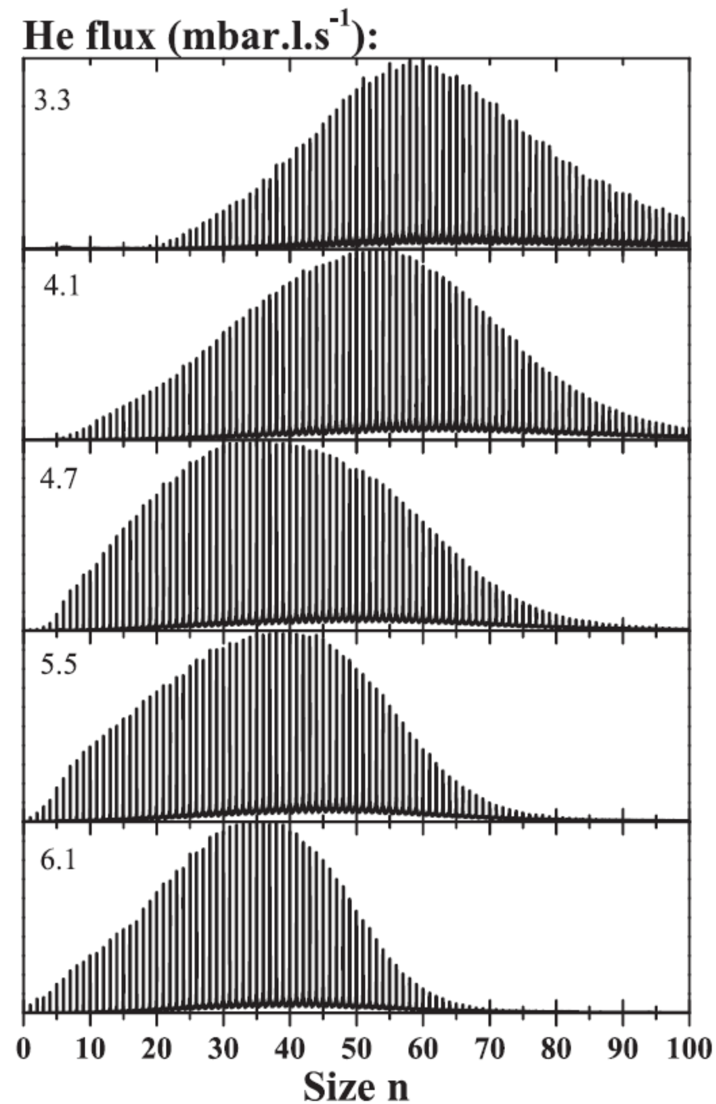
Influence of helium flux

Less time to grow as the flux is increased

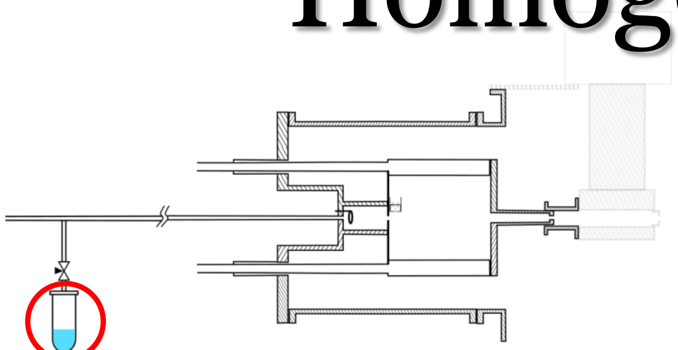
Discharge ionization

P(H_2O) = 23 mbar @ 293K

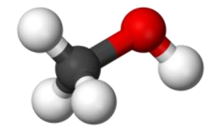
~ 1 % water in helium



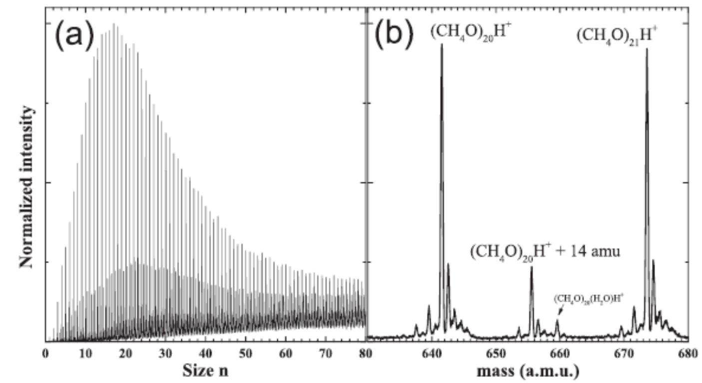
Homogeneous clusters



Methanol:



CH_4O



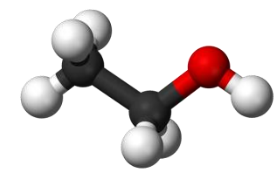
Clusters of relatively high vapor pressure materials can be produced the same way

P(Methanol) = 129 mbar @ 293K

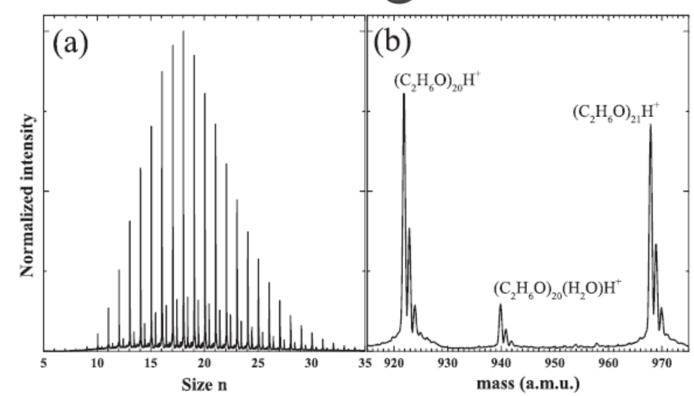
P(Ethanol) = 59 mbar @ 293K

Electron gun ionization

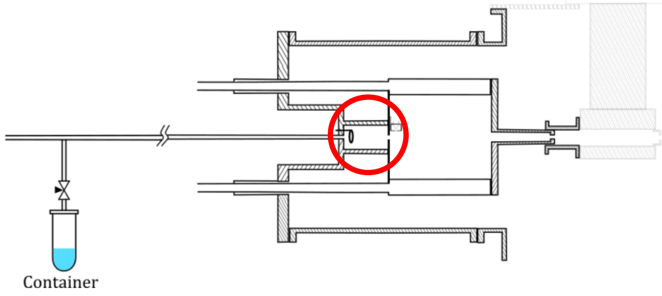
Ethanol:



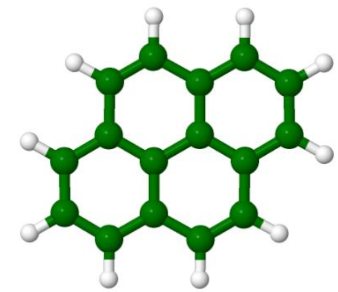
C_2H_6O



Homogeneous clusters



$C_{16}H_{10}$



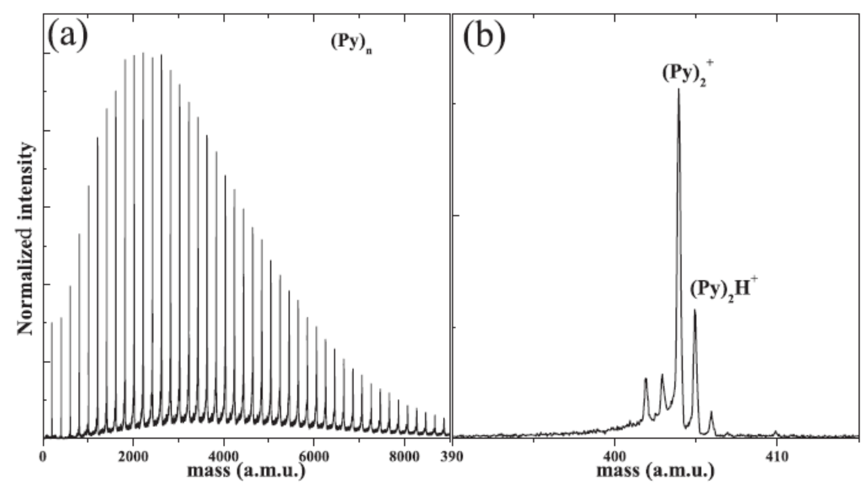
Clusters of relatively low vapor pressure produced using the oven.

Ex: Pyrene

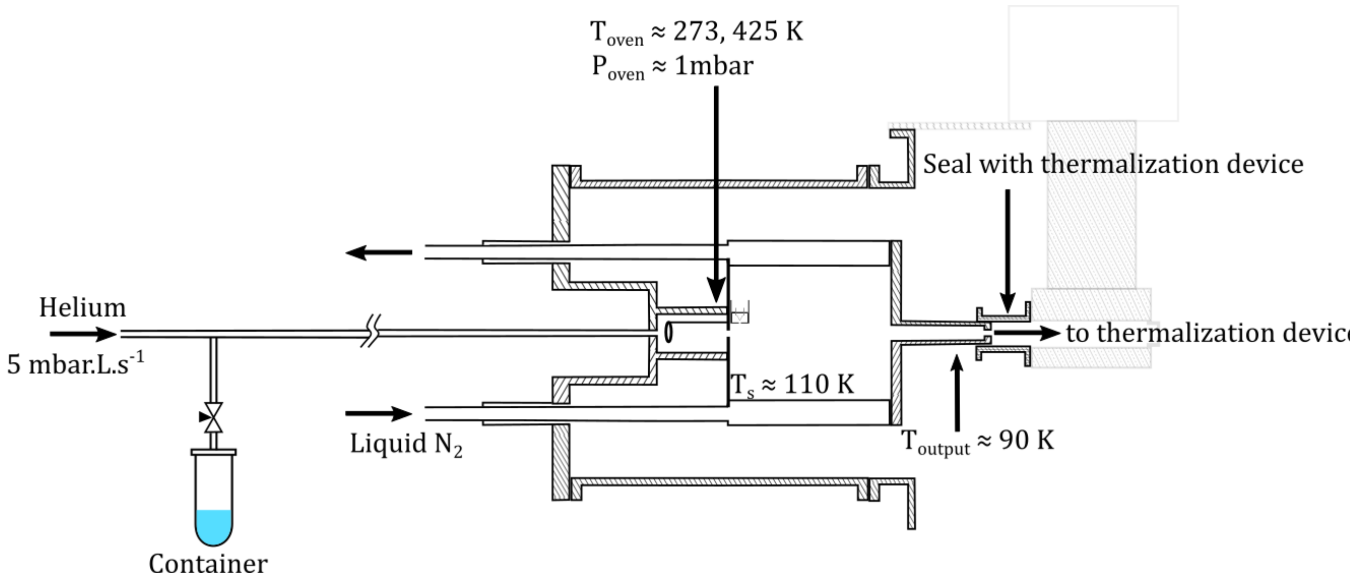
96%, powder

Electron gun ionization

$P(C_{16}H_{10}) = 5.10^{-4}$ mbar @ 315 K



Performances



Stable production for several hours

Need for regular (but not so frequent) cleaning (discharge, electron gun)

Current ~ 10^{11} ions/s (~ 20 nA)

Positive and negative ions

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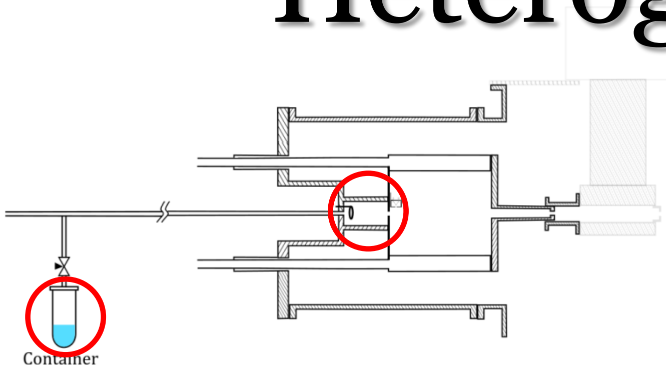
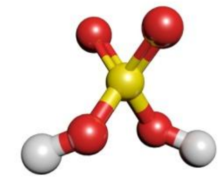
Heterogeneous clusters



H_2SO_4

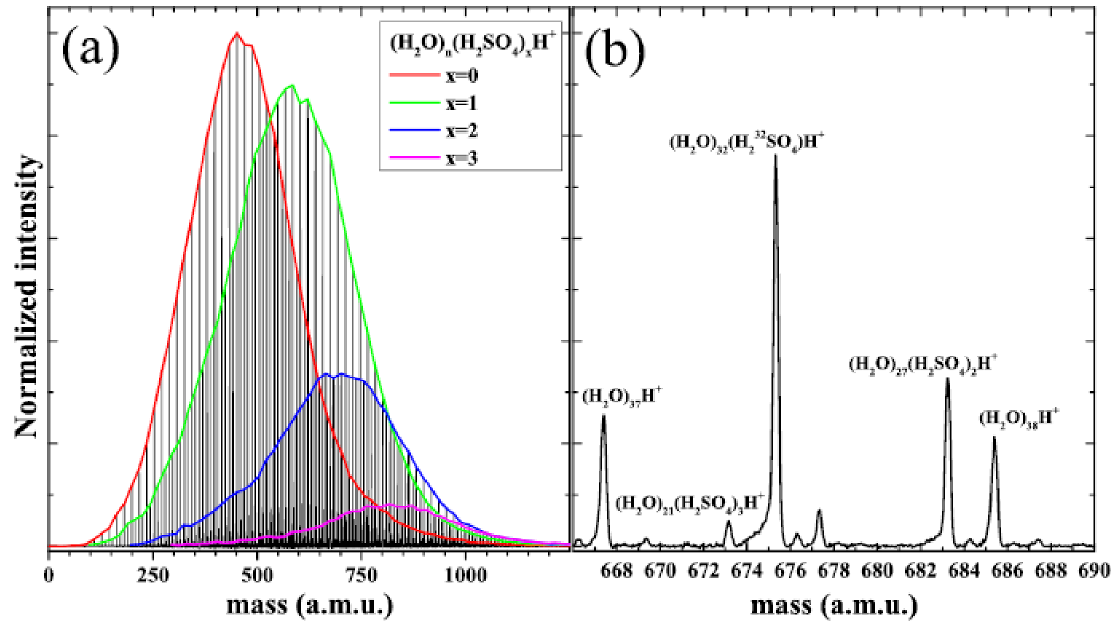


Ex: Water + sulfuric acid

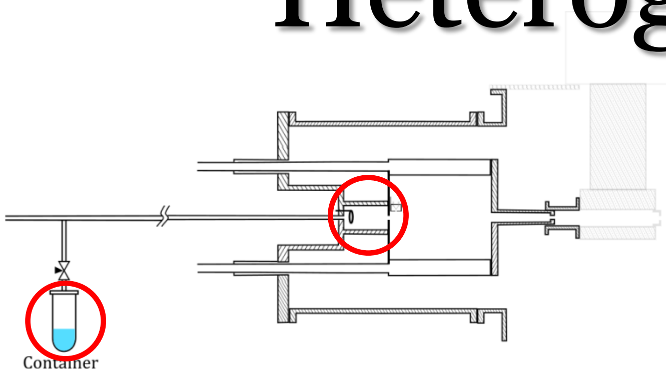
Discharge

99.99% liquid H_2SO_4

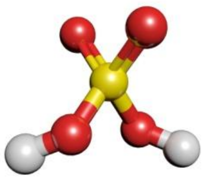
$P(H_2SO_4) = 5.10^{-4}$ @ 323 K



Heterogeneous clusters



H_2SO_4



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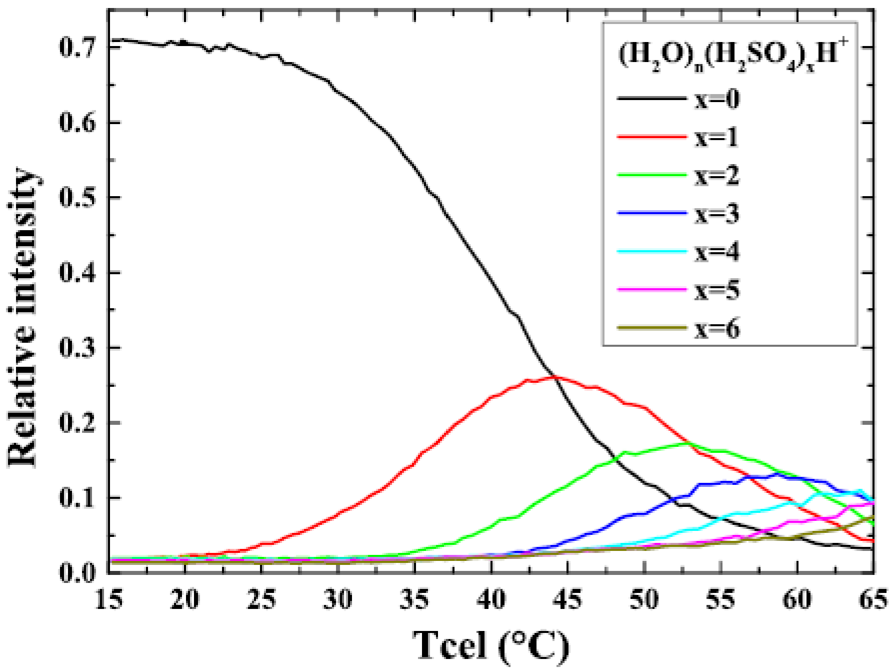
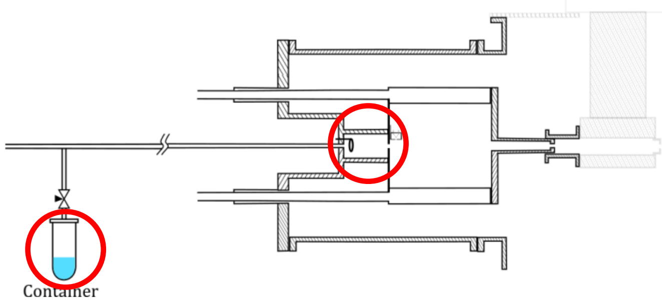
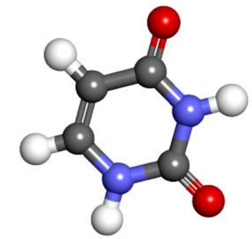


Heterogeneous clusters



$C_4H_4N_2O_2$

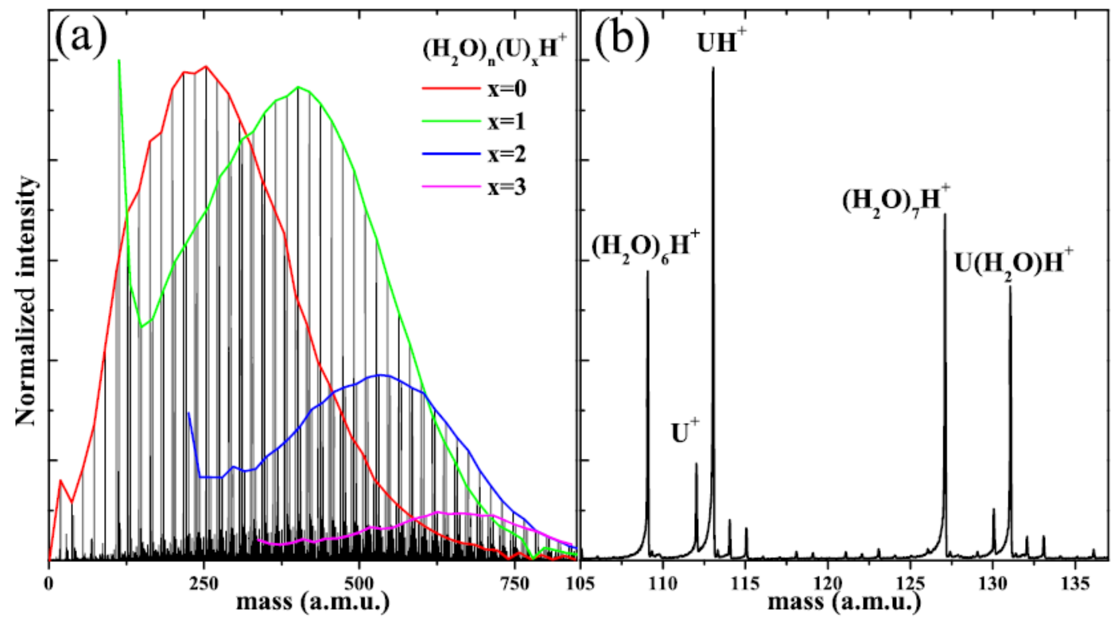


Ex: Water + Uracil

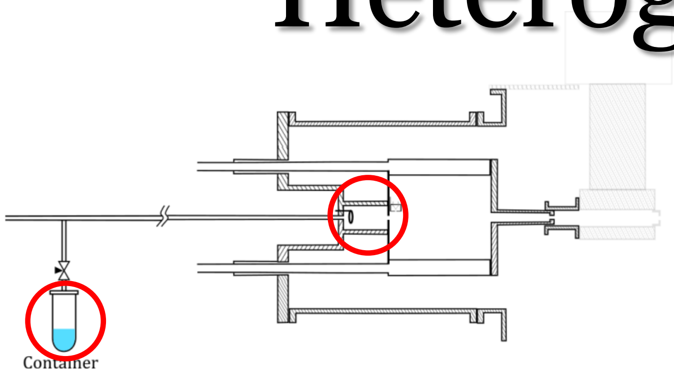
Discharge

99% Uracil powder

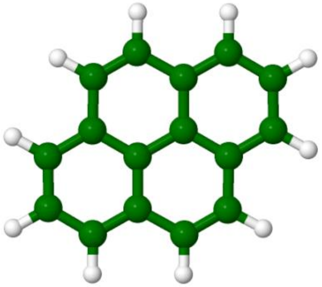
$P(C_4H_4N_2O_2) = 7.10^{-2}$ @ 378 K



Heterogeneous clusters



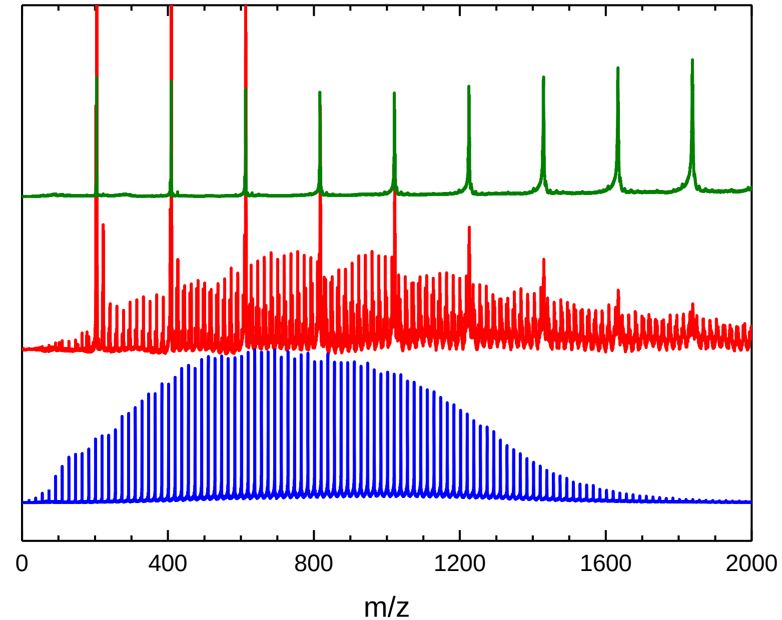
$C_{16}H_{10}$



Ex: Water + Pyrene

Discharge

T_{oven} ~ 323 K



$(Py)_n^+$

$(Py)_n(H_2O)_m^+$

$(H_2O)_mH^+$

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Conclusion

Cons:

Cluster formation not very well understood

Long term stability

Need regular cleaning

Pros:

Easy cluster size tuning

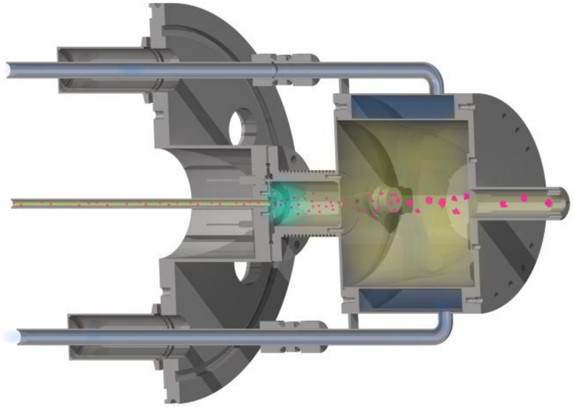
Continuous cluster production

Allow cluster thermalization

Source can be easily adapted to different species (even corrosive)

Very different vapor pressure compounds can be mixed

Thanks



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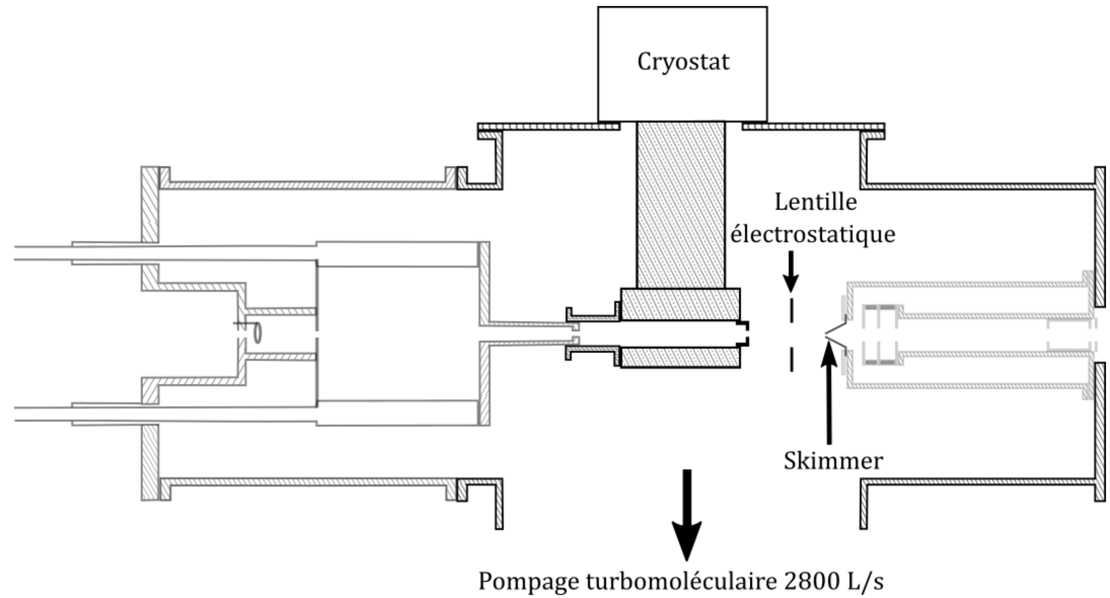
Gas aggregation source



Thermalization, T = 25 to 300 K

→ ~ 10^4 collisions, P ~ 1 mbar

→ Vacuum chamber P ~10^{-4} mbar

→ After skimmer P ~ 10^{-6} mbar

I. Braud *et al*, Rev. Sci. Instrum. **88**, 043102 (2017)