DIAGNOSTICS AND MODELING OF COLD LABORATORY PLASMAS WITH HIGH HYDROGEN CONTENT. APPLICATIONS TO MOLECULAR ASTROPHYSICS

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Abstract

- Ions and transient species of astrophysical interest can be produced by cold plasmas in low pressure discharges for their spectroscopic and kinetic characterization, allowing to identify them in space and to unravel likely pathways of generation and destruction at interstellar conditions.
- NH_4^+ and ArH^+ formations in hollow cathode discharges are studied experimentally and theoretically in this work [1,2]. The dependencies of ion densities on electron temperature, gas pressure and relative concentrations of the precursors are characterized. The kinetic models include electron impact ionizations and dissociations, gas phase barrierless ion-molecule reactions and surface processes, and reproduce reasonably well the observed results.
- These plasmas have been used to measure, with more precision and number of lines than previously, the high resolution vib-rotational spectra of the ν_4 band of NH_3D^+ [3], favoring its identification in space [4]; and of the ν=1-0 band of $^{36}ArH^+$ and $^{38}ArH^+$ ions [5], found in the interstellar media (ISM) recently [6,7].

Cold Laboratory Plasmas *vs.* ISM Clouds

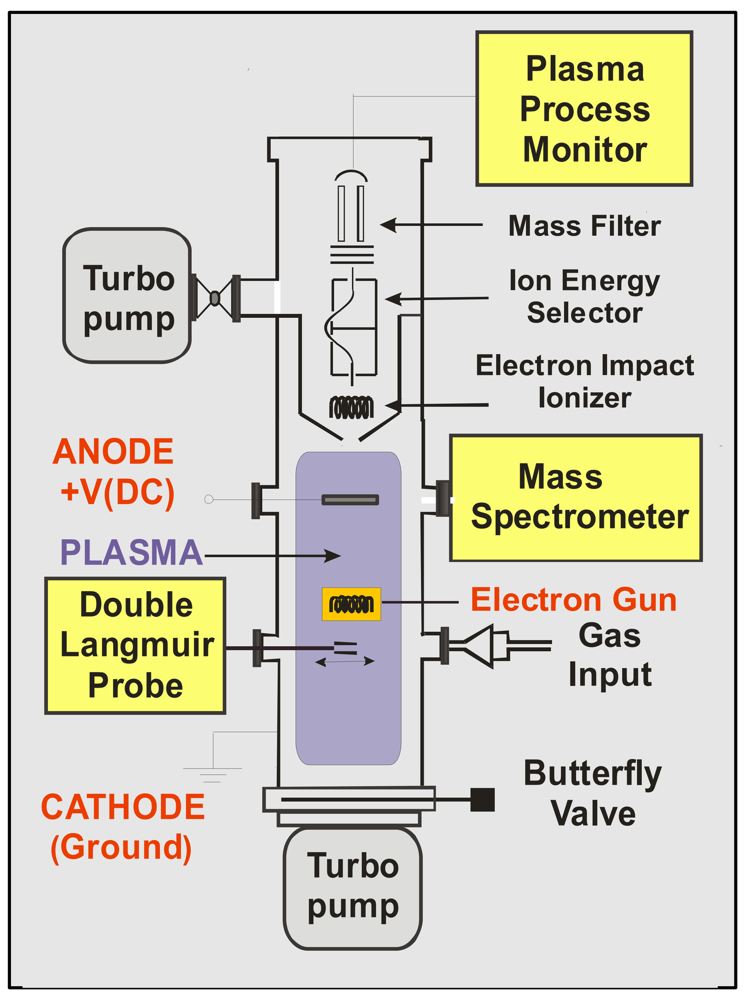
SIMILARITIES

- Gas-phase dominated by H_2
- Low ionization degree
- Low density (only binary collisions)
- Extensive ion-molecule chemistry (large rate coefficients ≠ f(T_{gas}))
- Surface chemistry necessary to explain molecular formation

DIFFERENCES

- Ionization and dissociation agents:
  Lab: electron impact *vs.* ISM : cosmic rays (& UV)
- Neutralizations of ions :
  Lab: neutralization in the walls vs. ISM : dissociative e^- attachment
- Surface chemistry: Different relevance of Eley-Rideal vs. Langmuir-Hinshelwood processes, depending on surface coverage.

Set-up for Plasma Kinetic Diagnostics



PLASMA GENERATION

Hollow cathode discharge DC reactor [1,2].

P = 0.8-8 Pa (10^{-14}–10^{-15} cm^{-3}). $t_{resid.} \cong 1$ s

$T_{gas} \cong T_{rotational} \cong 350$ K

$T_e \cong 2.8 - 4$ eV, $N_e \cong 10^{10}$-10^{11} cm^{-3}

DIAGNOSTIC TECHNIQUES

Quadrupole mass spectrometry:

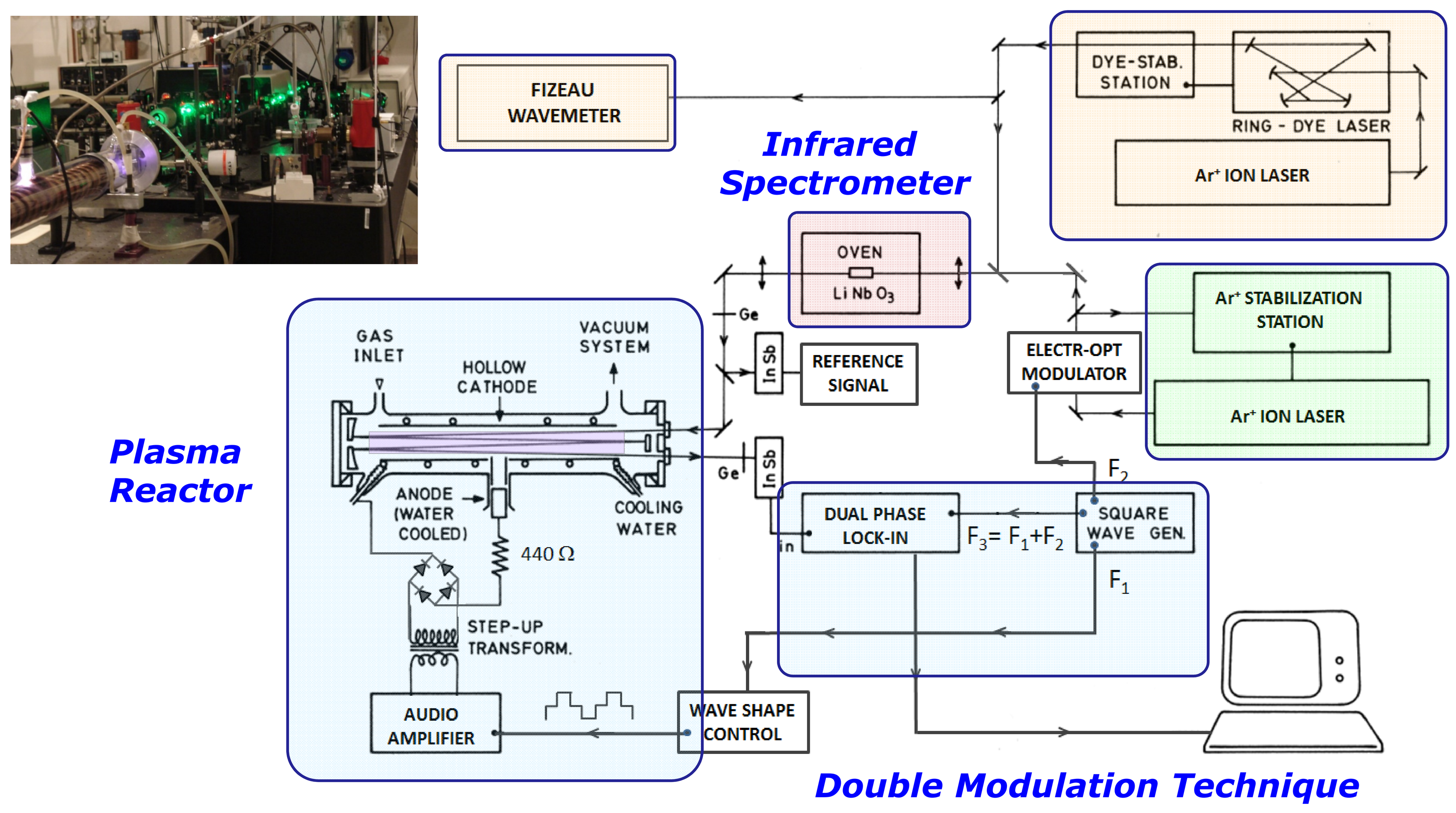
- For ions, with ion energy resolution.
- For stable neutrals.

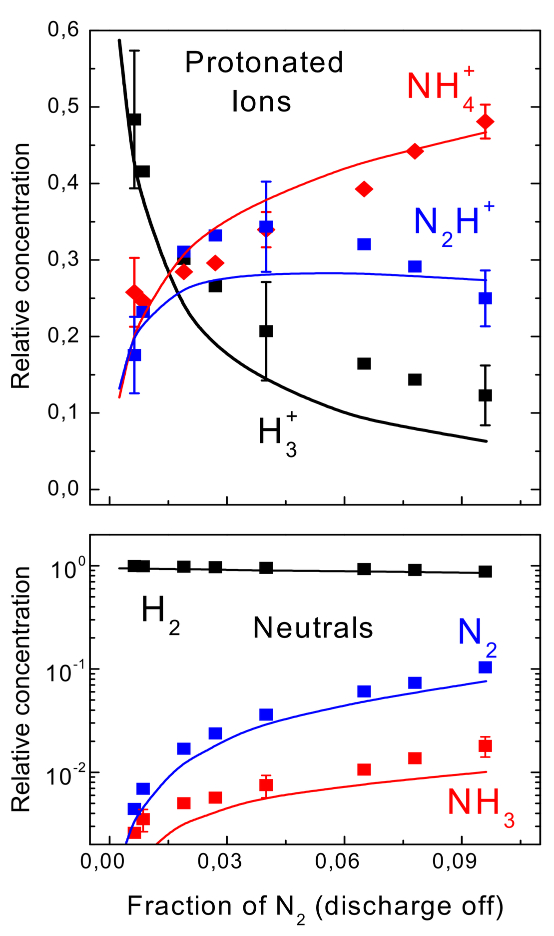
Visible emission spectroscopy:

- For excited species.

Double Langmuir probe: For charge density, N_e, and electron temperature, T_e.

Set-up for IR Difference-Frequency Laser Spectroscopy of Ions





N_2+H_2 Plasma Kinetics: Prevalence of NH_4^+

NH_4^+ is derived from the small amount of NH_3 produced at the reactor walls, and tends to prevail in the ion distributions even for NH_3 densities < 1%.

Main protonation reactions:

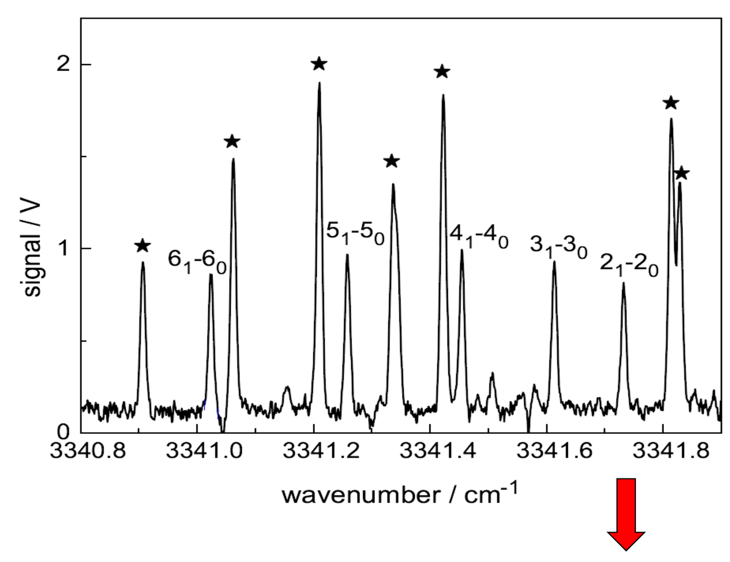
$H_2^+ + H_2 \rightarrow H_3^+ + H$ $H_3^+ + N_2 \rightarrow N_2H^+ + H_2$

$H_3^+ + NH_3 \rightarrow \mathbf{NH_4^+} + H_2$ $N_2H^+ + NH_3 \rightarrow \mathbf{NH_4^+} + N_2$

Evolution of the relative concentrations of neutral molecules and protonated ions as a function of the initial fraction of N_2 in H_2/N_2 discharge mixtures. Symbols: Experiment. Lines: Model.

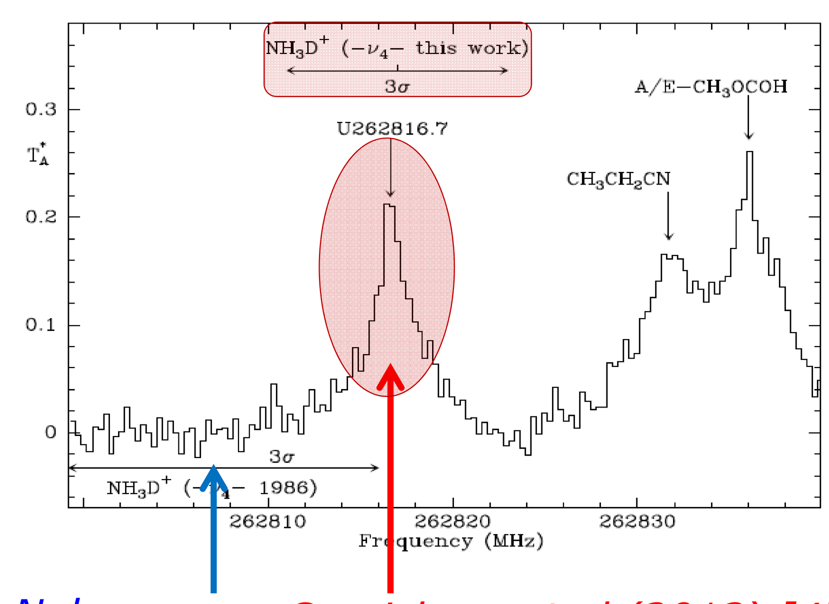
High Resolution IR Spectrum of NH_3D^+

IR spectrum of NH_3D^+ and $*NH_4^+$



New Predicted J_K (1_0-0_0) 262817 ± 6 MHz (±3σ)

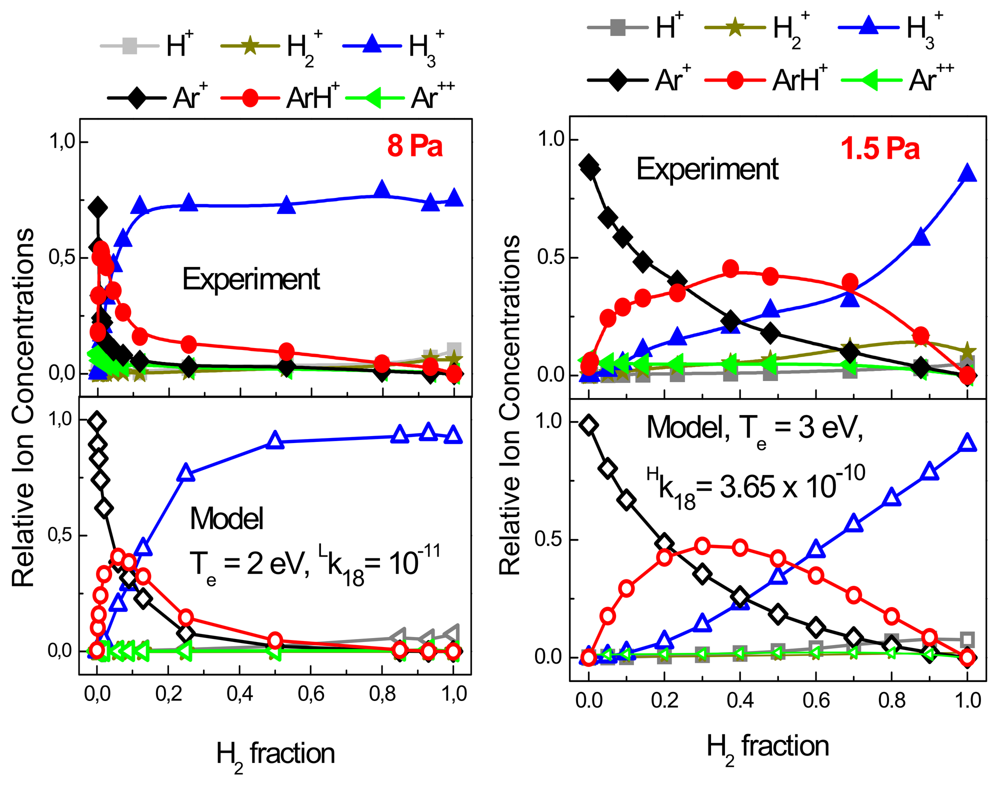
The new predicted frequency supports the identification of NH_3D^+ in the ISM



Nakanaga & Amano (1987)

Cernicharo et al (2013) [4]
Domenech et al (2013) [3]

$Ar+H_2$ Plasma Kinetics: Prevalence of ArH^+



- *Ion concentrations studied in $Ar+H_2$ discharges at 8 and 1.5 Pa with different Ar/H_2 ratios.*
- *ArH^+ ions concentration depends strongly of electron temperature and of H_3^+ vibrational excitation.*

Key reactions:

ArH^+ Formation

1) $e^- + Ar \rightarrow Ar^+ + 2e^-$

2) $Ar^+ + H_2 \rightarrow ArH^+ + H$

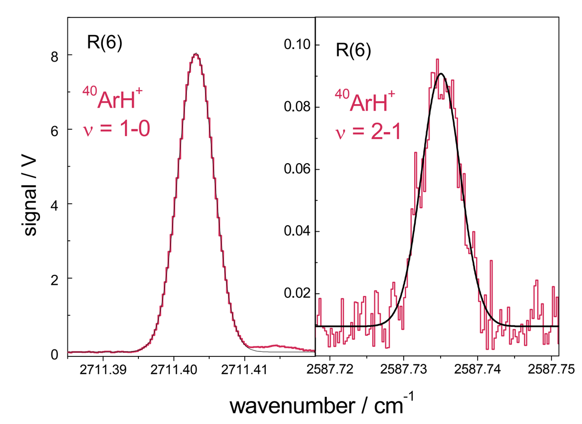
ArH^+ Destruction

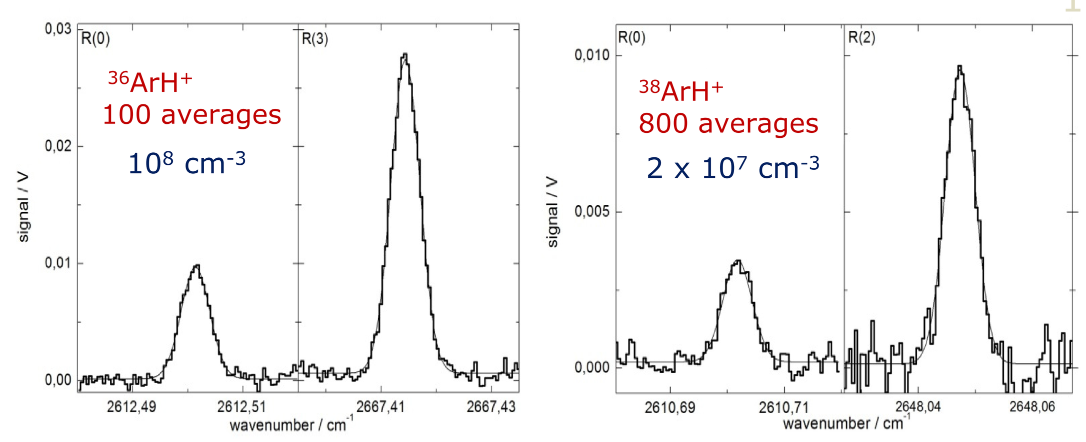
$ArH^+ + H_2 \rightleftharpoons H_3^+(v) + Ar$

High resolution IR spectra of $^{36}ArH^+$ & $^{38}ArH^+$

Solar wind composition: ^{40}**Ar**/ ^{38}Ar/ ^{36}**Ar** = **0.0** / 15.0 / **85.0** %

Earth's atmosphere compos.: ^{40}**Ar**/ ^{38}Ar/ ^{36}**Ar** = **99.6** / 0.06 / **0.34** %





$^{40}ArH^+$:
S/N ~1000 (ν = 1-0) 1 scan.
[$^{40}ArH^+$] ~ 4 x 10^{10} cm^{-3}
Gas temperature: 390 ± 10 K

19 lines measured. Many, for the first time. Used in a Dunham-type global fit of all published IR and sub-mm laboratory data of all isotopologues [5].

References

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