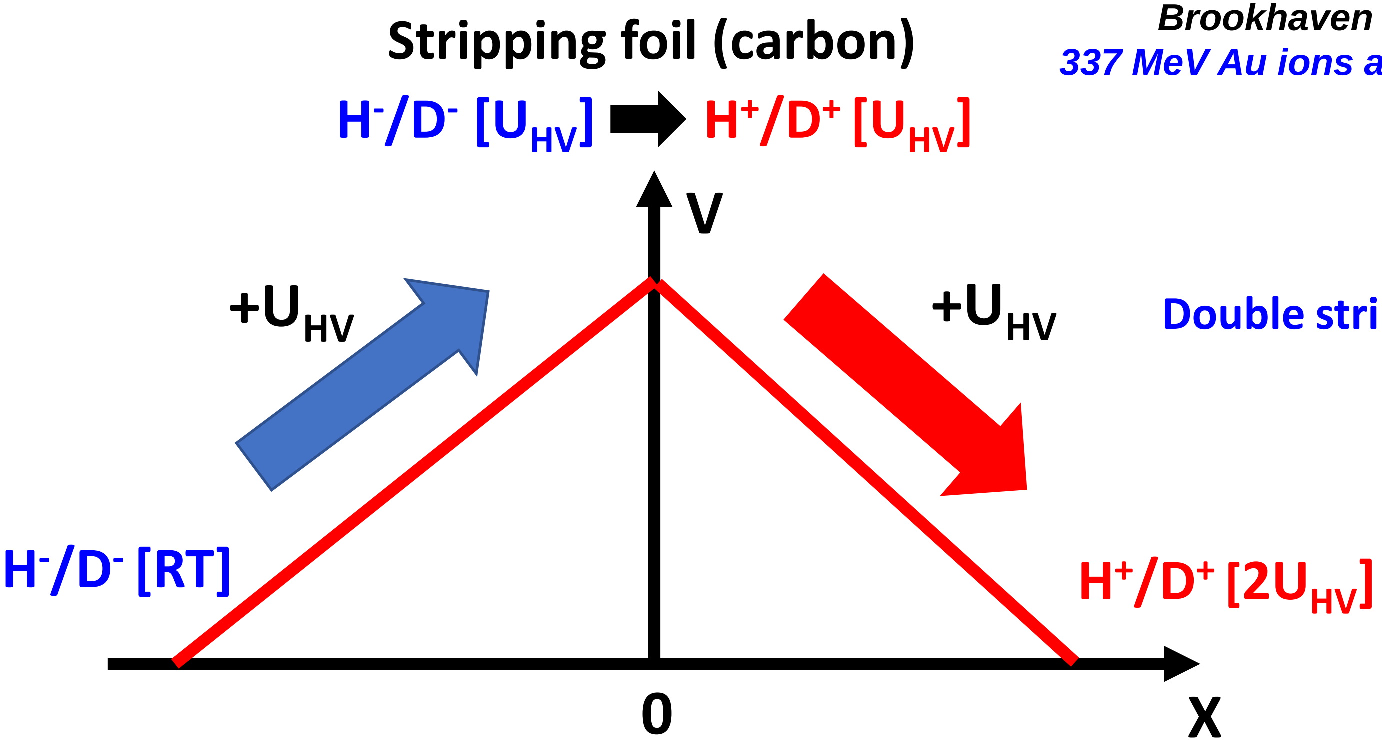
Physics of negative hydrogen ion sources: a review

L. Schiesko

Historical need of H^-/D^- sources

- **Van de Graaf and Tandem accelerators**
- **Double the acceleration voltage with one HV terminal**
- **Removal of 2 e^- is simple !**

Brookhaven National Laboratory
337 MeV Au ions and any other kind of ions



Double stripping of negative ions

L. Alvarez, Rev . Sci. Inst. 22, 705 (1951)
G. I. Budker and G. I. Dimov, in Trudy Mezhdunarodnoi Konf. Po Uskoritelyam (1963)
G. I. Budker, G. I. Dimov, and V. G. Dudnikov, Atomnaya Energiya 22, 348 (1967)

Main applications of H^-/D^- sources

- 1960's used in cyclotron and charge-exchange was used for cyclic high-energy proton accelerators

G. I. Budker and G. I. Dimov, in Proceedings of the Conference on High Energy Accelerators, 993 (1963)
K. W. Ehlers, Nucl. Instrum. Methods 32, 309 (1965)
O. Felden et al., AIP Conf. Procs. 1515 321 (2013)

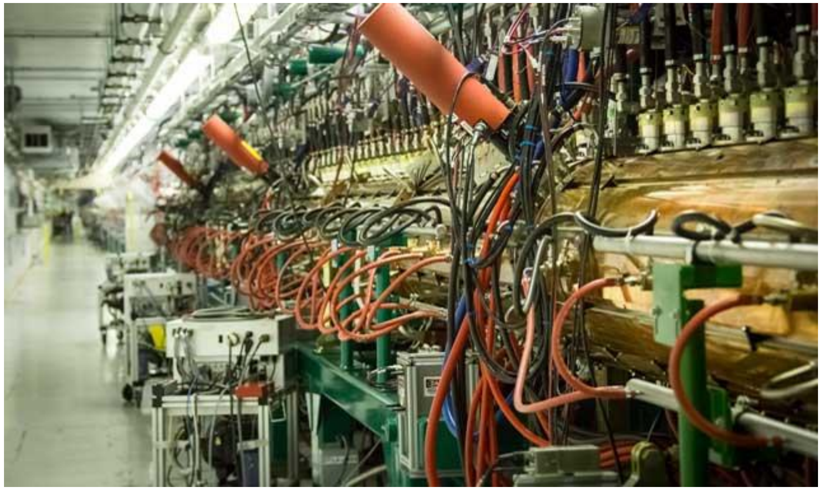
- 1970's H^- / D^- for to inject H^0 / D^0 with $E > 100$ keV / nucleons: injection into magnetic fusion device (1 MeV required for ITER)

K. H. Berkner et l., Nucl. Fusion 15 249 (1975)
M. Bacal and M. Wada, Plasma Sources Sci. Technol. 2 190309 (2020)

- Neutron sources: spallation neutron source

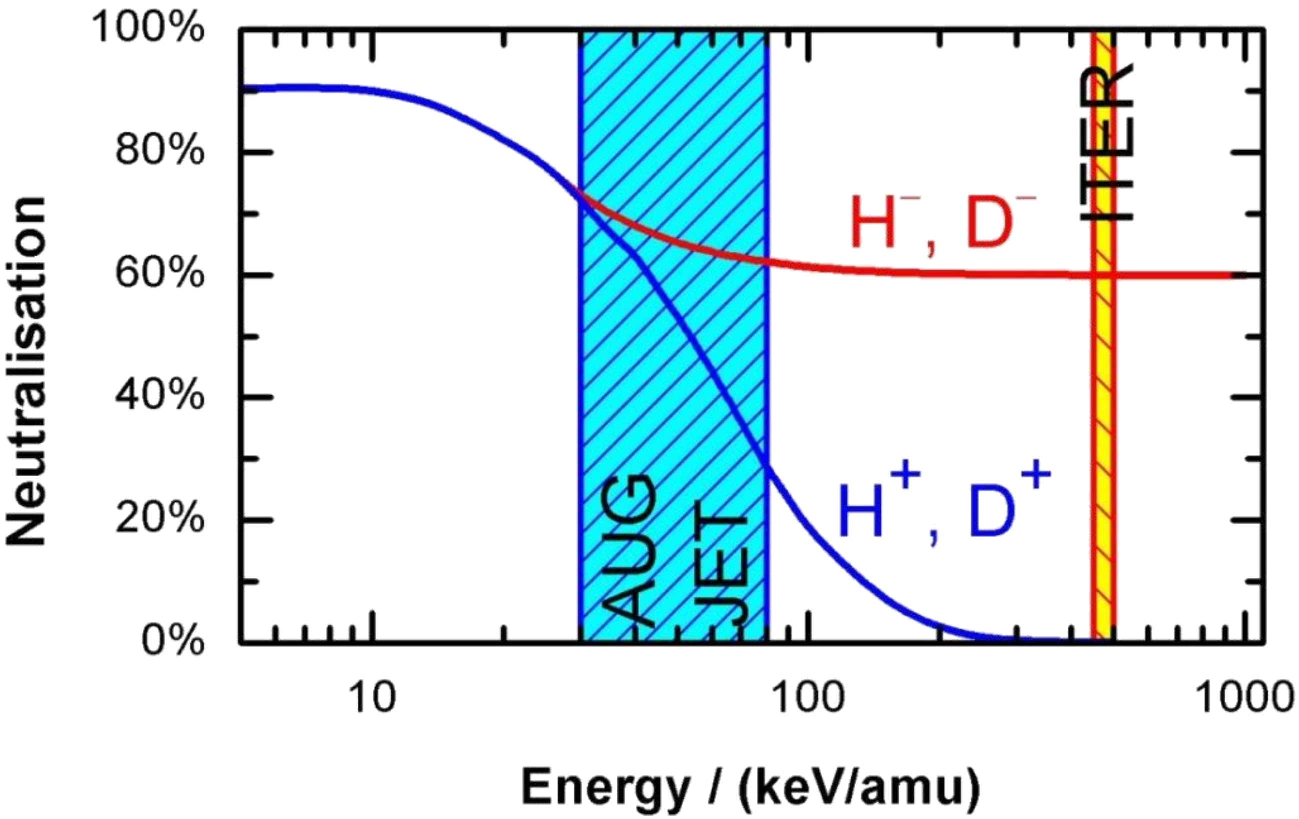
M. P. Stockli et al., AIP Conf. Procs. 1869 030010 (2017)

H^-/D^- production much more difficult than H^+/D^+


Oak Ridge National Laboratory

Do we really need negative ions ?

- Proton generation very simple and effective, not negative ions !
- Example: ITER Neutral Beam Injectors requires 1 MeV H^0 or 870 keV D^0 for plasma heating



- Neutralisation efficiency by stripping of positive ions too small
- Example: particle accelerators, double charge-exchange doubles the acceleration energy

Negative ions generated in a low temp. plasma source

Content of this talk

- **Fundamental mechanisms governing low pressure, low temperature H^-/D^- sources**
  - **Volume enhanced mechanisms**
  - **Surface conversion mechanisms**
  - **Typical setup and comparison of sources**

- **Mostly used source diagnostics**
  - **Amperemeters, Langmuir probes, OES**
  - **Laser detachement, cavity ringdown spectroscopy, Fowler method**

- **Theory and modelling**
  - **0D, 1D Fluid and kinetic, 2D and 3D Particle-in-cell**

- **Beam physics and beam properties: out of the scope but questions are welcome**

Outline

I. Work principle

   i. Volume negative ions generation and destruction mechanisms, example of source

   ii. Surface negative ions generation, example of source

II. Diagnostics

III. Theory and modelling

IV. Conclusion

Outline

Volume generation mechanism

Main generation mechanism: two steps process

$$\mathrm{H_2(0,0) + e \xrightarrow{1} H_2(v',J) + e \xrightarrow{2} H + H^-}$$

1. Vibrational excitation of H_2 / D_2 by electron impact

2. Dissociative electron attachement to a ro-vibrationally excited H_2 / D_2

Volume effect: M. Bacal, Phys. Scripta 2/2 467 (1982)

Other mechanisms

$$\mathrm{H + e \rightarrow H^- + h\nu}$$
$$\mathrm{H_2^+ + e \rightarrow H^- + H^+}$$
$$\mathrm{H_3^+ + e \rightarrow H^- + H_2^+}$$

$$\mathrm{H^+ + H_2 \rightarrow H^- + H_2^+}$$
$$\mathrm{H_2^+ + H_2 \rightarrow H^- + H + 2\,H^+}$$
$$\mathrm{H_2^+ + H_2 \rightarrow H^- + H^+ + H_2^+}$$

Smaller cross-section, minority plasma molecular ions

M. Bacal et al., J. Appl. Phys. 129, 221101 (2021)

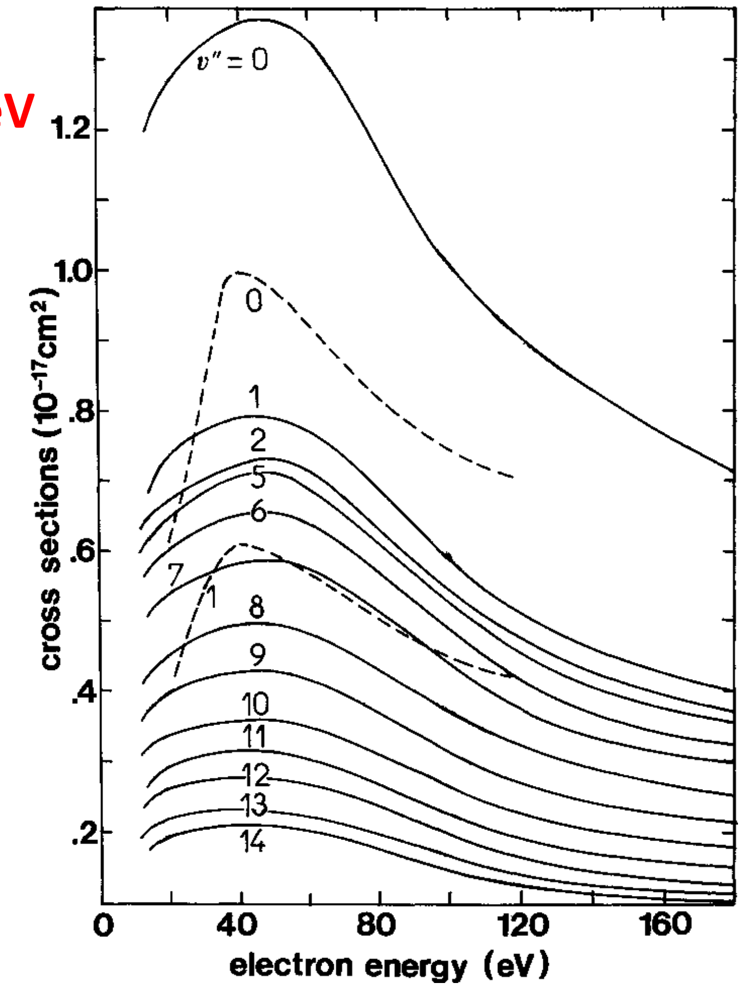
T. J. Millar, C. Walsh, and T. A. Field, Chem. Rev. 117, 1765–1795 (2017)

Excitation of hydrogen molecules

1. Vibrational excitation of H_2 / D_2 by electron impact

$$e + H_2 / D_2(0,0) \rightarrow H_2 / D_2(v', J) + e$$

- Threshold and max. around 20 eV and 40 eV repectively
- High T_e required
- Typical neutral H_2 background pressure 0.1 - 10 mTorr
- Collisions of H_2 / and recombinative desorption adsorbed atoms from the walls also yield $H_2 / D_2(v', J)$



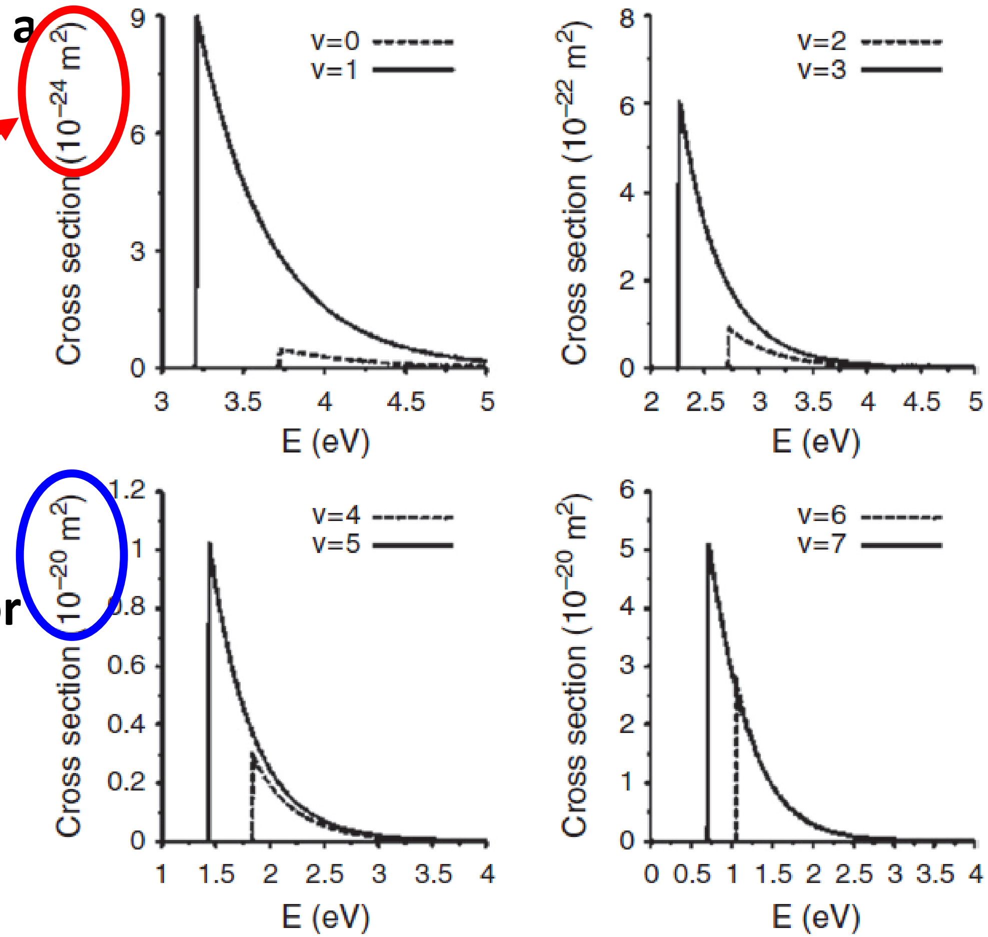
J. Marx et al., Journ. de Physique 46 1667 (1985)

Dissociative electron attachement (DEA)

2. **Dissociative electron attachement to a ro-vibrationally excited H_2 / D_2**

$$\mathbf{H_2(v', J) + e \rightarrow H + H^-}$$

- **Cross section very small for v' = 0**
- **Gain of several orders of magnitude for v' > 1**
- **Low energy electrons needed for DEA attachement !**



J. Horacek et al., Phys. Rev. A 70, 052712 (2004)

Main destruction mechanisms

- H electron affinity is **0.7542 eV**

- H^- / D^- **easily destroyed** in low temperature plasma volume:

[1] $$H^- + e \rightarrow H + 2e \propto T_e$$
$$H^- + H^+ (H_2^+ \text{ or } H_3^+) \rightarrow 2H \propto T^+$$
$$\left.\begin{array}{l} H^- + H \rightarrow 2H + e \\ H^- + H \rightarrow H_2 + e \end{array}\right\} \propto T_H$$

R. K. Janev et al., Elementary Processes in Hydrogen-Helium Plasmas (1985)

C. Sidis et al., Phys. Rev. Lett. 47, 1280 (1981)

- [1] negligible for $T_e < 2$ eV: use of magnetic field (MF $\propto$ 10 mT) to reduce T_e

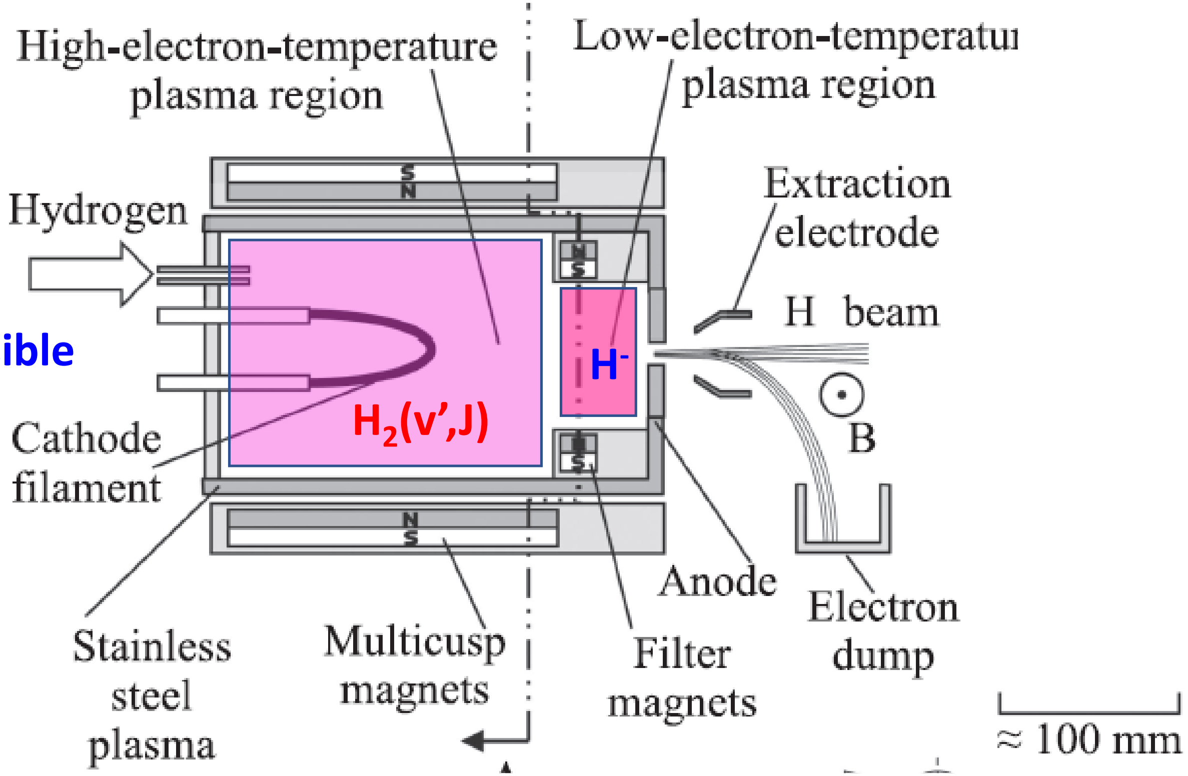
- **Magnetic field**
  - Increase of H^- volume generation (source dep.)
  - Decreases the H^- destruction by e^- collision
  - Reduces the amount of co-extracted electrons

Example of volume source setup

- **High and low energy electrons: $H_2(v',J)$ and electron attachement / minimization of H^- destruction and co-extracted electrons**

- **Use of a magnetic field with a typical induction of 10 mT**

RF possible



D. Faircloth et al., New J. Phys. 20 025007 (2018)

- **Typical parameters: p = 0.1 - 10 mTorr, 10^{15-17} m^{-3} positive ion density, T_e between 1 and 10 eV, kW range power and up to tens of kV for beam**

- **Performance of tens of $mA.cm^{-2}$ for the beam**

Outline

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Surface generation mechanism

Negative surface ionization

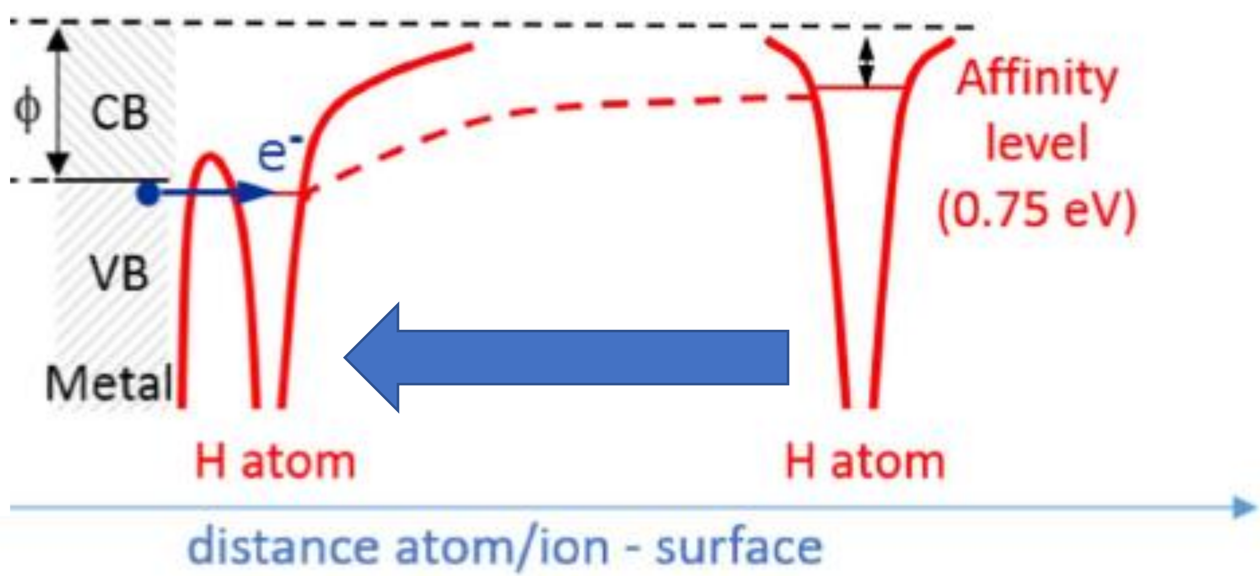
Fundamental level

Vacuum level

Conduction band

Fermi level

Valence band



G. Cartry et al., New J. Phys. 19 025010 (2017)

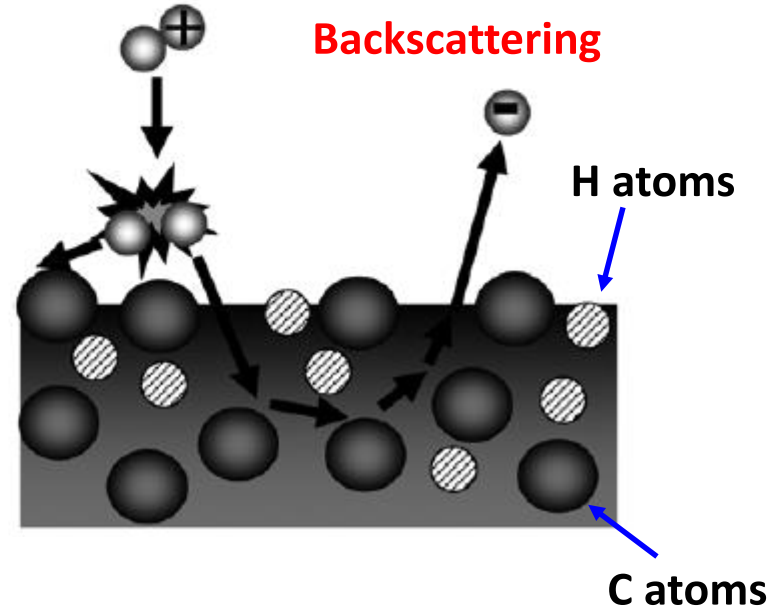
- **Image potential:** near surface downshift of H affinity level
- **Overlapping of surface VB and atom EA: electron tunneling possible**

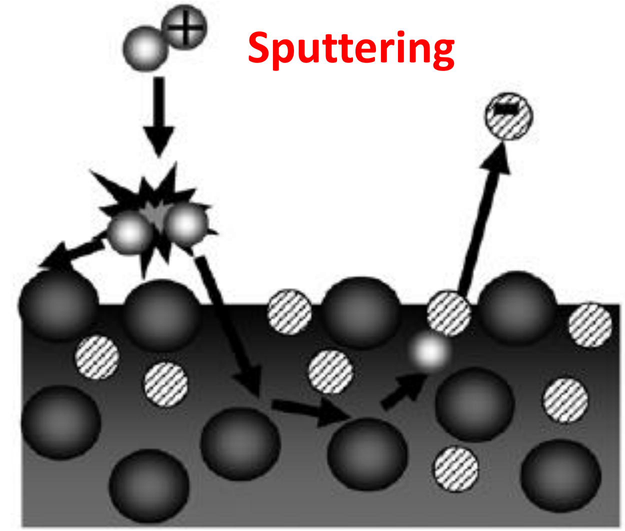
$$P^{-} \propto \exp[-(WF - EA)]$$

- **Back-tunneling is possible when leaving the surf. (neutralization)**
- **Electron affinity fixed to 0.75 eV for H, thus, low mat. WF must be used !**
- **Low mat WF around 2 to 3 eV (Cs, Ba, LaB6, K, Rb …)**

Surface atomic and molecular mechanisms

- **For plasma volume atomic species: negative surface ionization (backscattering):**
  - **single (H^0/D^0) or double electron capture (H$^+$/D$^+$)**
  - **fragmentation upon impact and negative ionization of molecular ions (H_x^+/D_x^+) with x = 2 or 3**



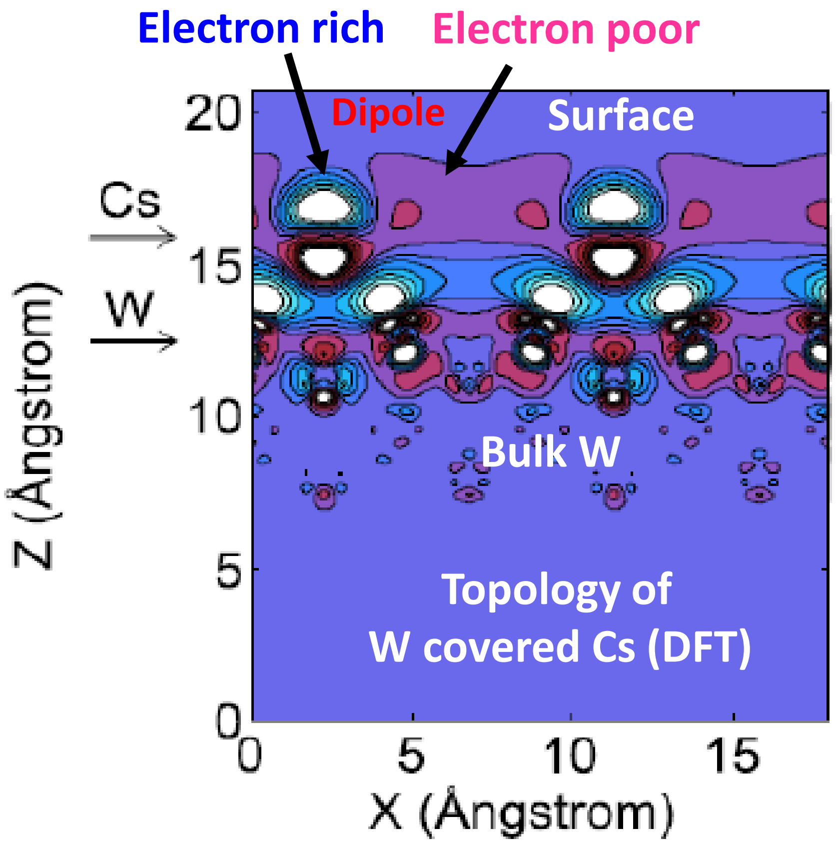


- **Sputtering followed by surface negative ionization of an adsorbed H or D atom by an incoming plasma ion (energy and surface temp. dependent)**

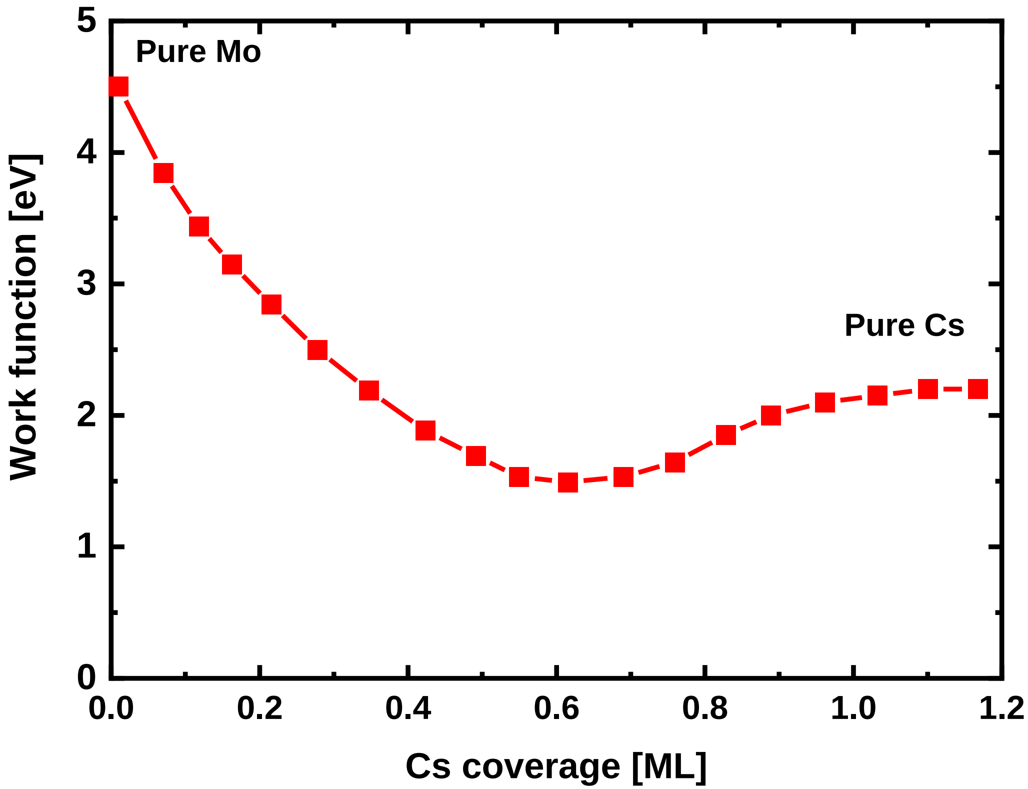
L. Schiesko et al., Plasma Sources Sci. Technol. 19 045016 (2010)

Low work function materials

- Alkali metal evaporated from an oven is performed, mostly Cs



S. Chou et al., J. Phys. Condens. Matter 24 445007 (2017)



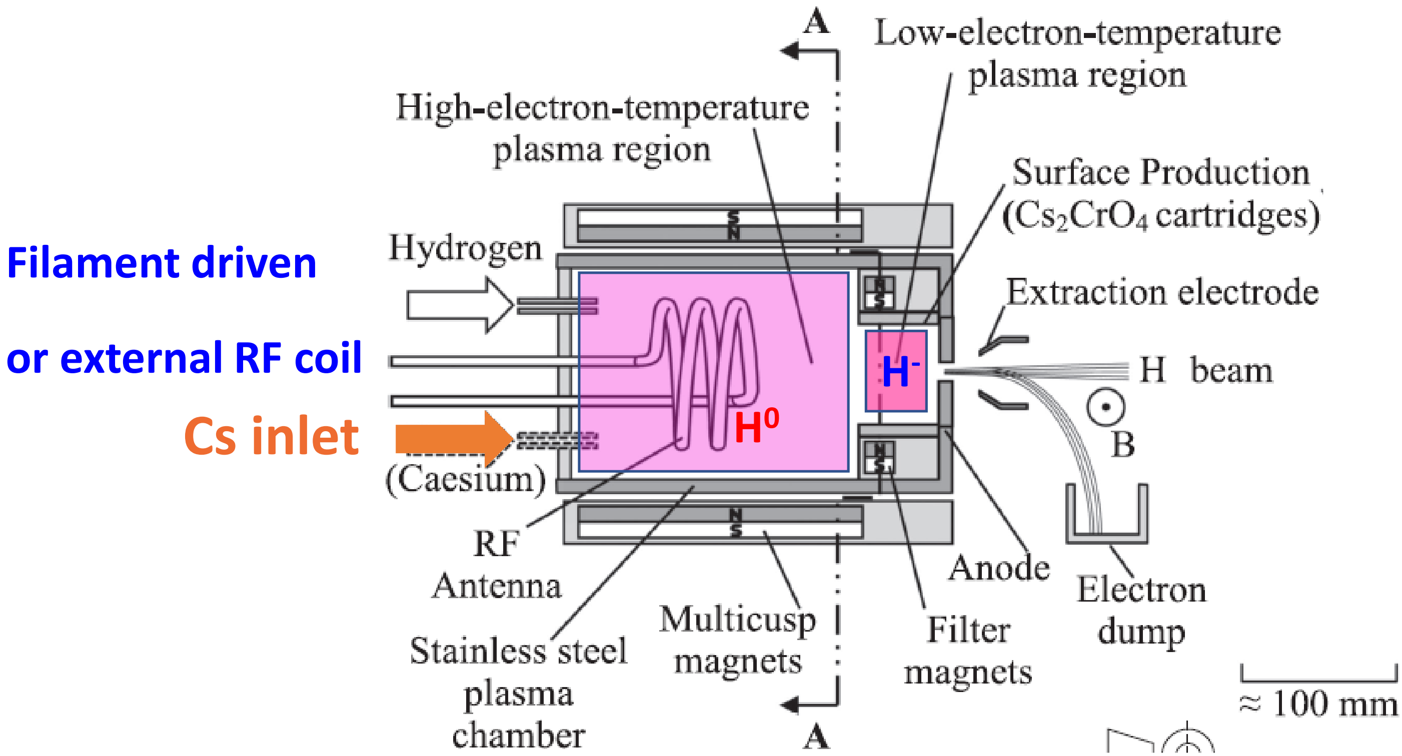
L. W. Swanson and R. W. Strayer, J. Chem. Phys. 48, 2421 (1968)

- Dipole effect and Cs-Cs orbital overlapping decreases the WF
- Strong reduction of the WF, on most of the material over which it is deposited
- Bulk Cs WF recovered for more than 1 mono-layer

Example of surface conversion source setup

- **High and low energy electrons: H_2 dissociation and minimization of H^- destruction and co-extracted electrons, magnetic field 10 mT**

- **Cs evaporated from an oven, covers all the chamber surfaces**



D. Faircloth et al., New J. Phys. 20 025007 (2018)

- **Typical parameters: p = 0.1 - 10 mTorr, $10^{16\text{-}18}$ m^{-3} positive ion density, T_e between 1 and 20 eV, kW range power and up to tens of kV for beam**

- **Performance of tens of $mA.cm^{-2}$ for the beam**

Other sources

- **Magnetron and Penning surface plasma sources**
  - Compact magnetized & caesiated sources
  - Bright H^- sources: around 100 mA, cathode surface (-50 V)
  - Magnetron: direct extraction, Penning: after Cx with slow H^0

Penning: D. Faircloth et al., AIP Conf. Proc. 1390 205 (2011)
Magnetron: D. S. Bollinger, AIP Conf. Proc. 1655 070002 (2015)

- **RF-driven volume and surface sources**
  - Caesiated sources, relies on both surface and volume generation
  - Often pulsed sources, around 50 mA extracted current

J. Lettry et al., AIP Conf. Proc. 1869 030002 (2017)
M. P. Stockly et al., AIP Conf. Proc. 1869 030010 (2017)
M. Cavenago et al., Rev. Sci. Instrum. 91, 013316 (2020)

- **ECR, ICP and Helicon sources (extraction in progress)**

Cs free:
  - ECR: volume operated, $n_{H^-} \propto 6.10^{15}$ m^{-3} (1 kW)
  - ECR / ICP: strong surface conversion yields on Cs-free mats. (HOPG, BDD, Gd …)
  - Helicon: volume operated, $n_{H^-} \propto 10^{16}$ m^{-3} (up to 10 kW)

Cs free review: F. Taccogna et al., Eur. Phys. J. D 75 227 (2021)
Sources for acclerators review: D. Faircloth et al., New J. Phys. 20 025007 (2018)

ECR: S. Aleiferis et. al, Plasma Sources Sci. Technol. 27, 075015 (2018)
ECR / ICP: G. J. Smith et al., J. Phys. D: Appl. Phys. 53 465204 (2020)
Helicon: D. S. Bollinger, AIP Conf. Proc. 1655 070002 (2015)

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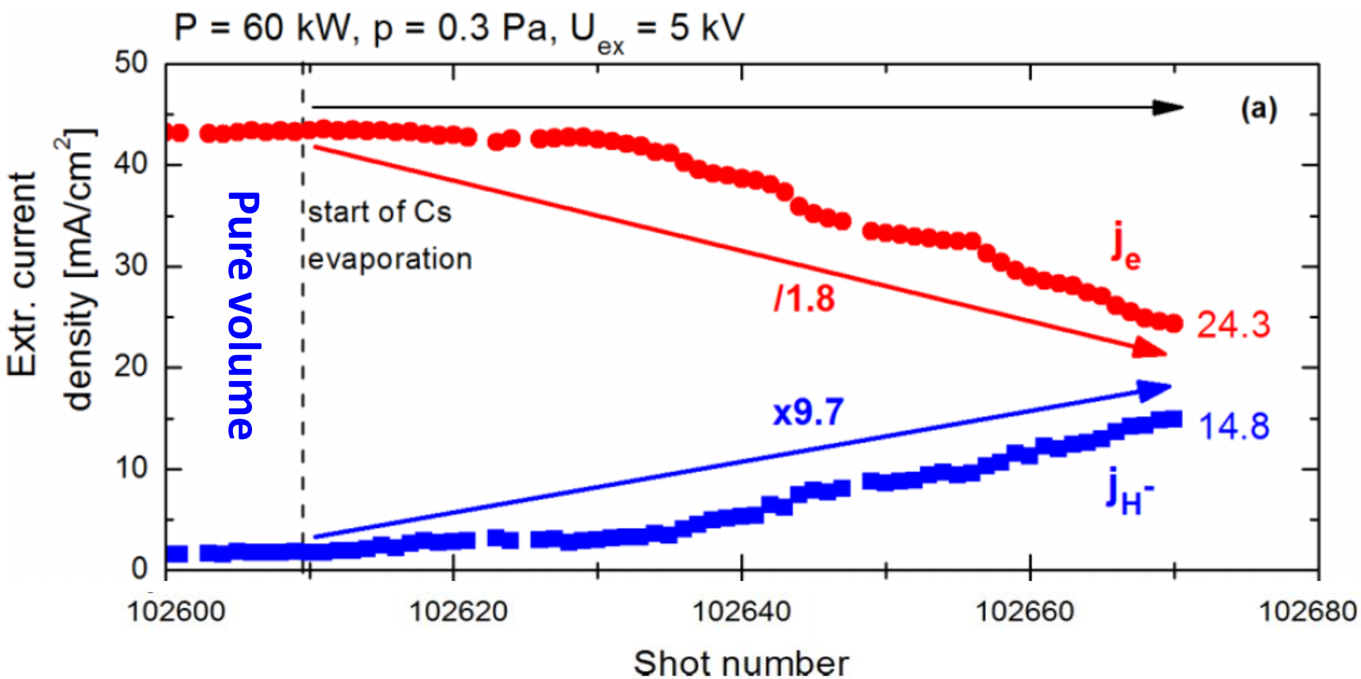
Source performance: amperemeters

Most important diagnostic for source performance !



C. Wimmer, L. Schiesko et al., Rev. Sci. Inst. 87 02B310 (2016)

- **Cs-free source: volume operation** ➡ **low H^-, high e^-**

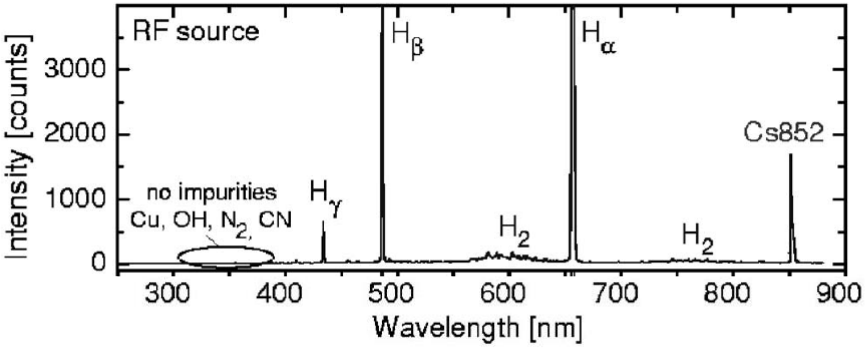
- **Cs evaporation: reduction of co-ext. e^- and strong increase of ext. H^-**

  « Cs effect »: Y. I. Belchenko et al., Nucl. Fusion 14, 113 (1974)

- **Cs management: control of the evaporation rate can be tricky !**

Optical Emission Spectroscopy (OES)

- **LOS integrated, non-invasive technique, absolute calibration**

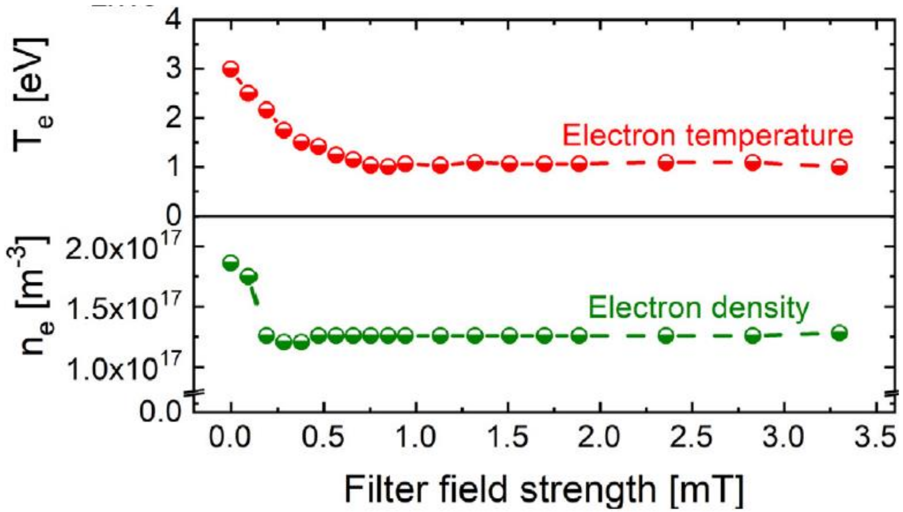


U. Fantz et al., Rev. Sci. Inst. 77 03A516 (2006)

Basics of OES: U. Fantz, Plas. Sources Sci. Techn. 15 S137 (2006)

- **Determination of n_e by H_β and H_γ line intensity-ratio (Balmer series), T_e coupling of OES to a collisional-radiative (CR) model**

D. Wünderlich et al., J. Quant. Spectrosc. Radiat. Transfer 110, 62 (2009)



D. Wünderlich et al., J. Quant. Spectrosc. Radiat. Transfer 240, 106695 (2020)

- **Atomic and molecular hydrogen density, neutral gas temperature: H_γ / Fulcher line intensity-ratio coupled to CR**

U. Fantz. et al., Nucl. Fusion 46 S297 (2006)

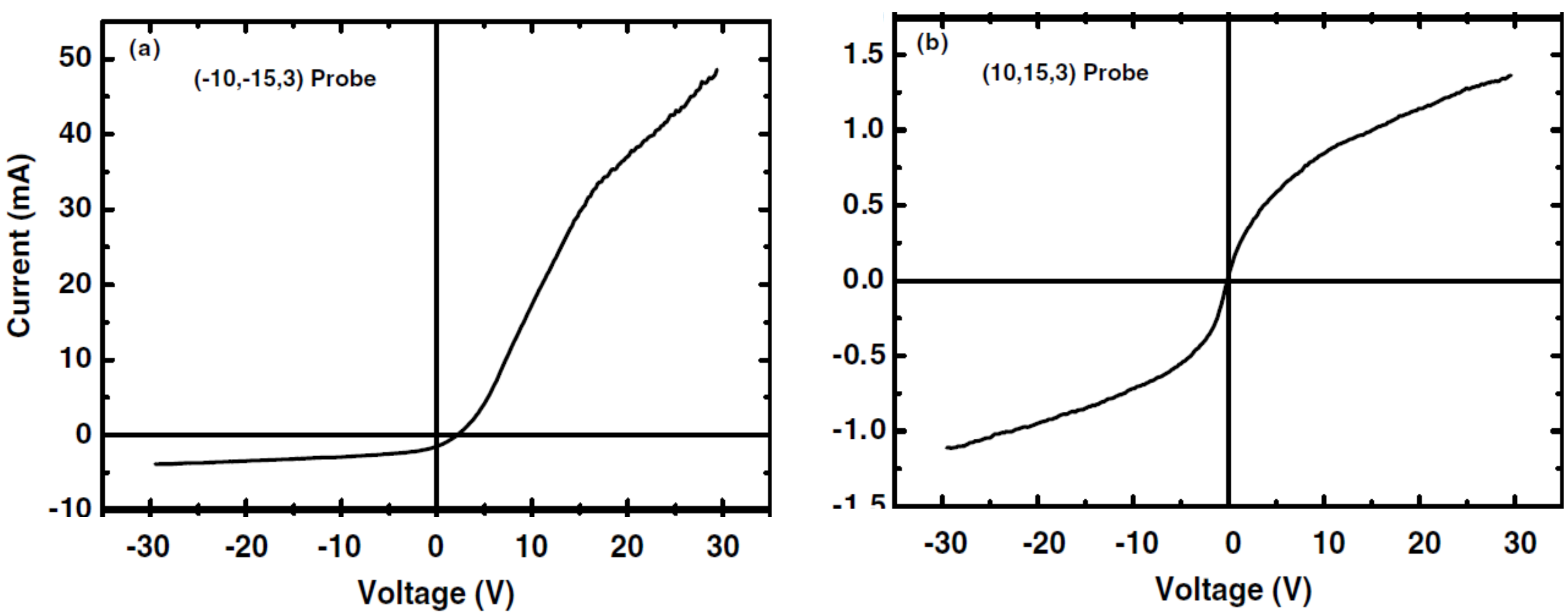
- **Monitoring of Cs and impurities (O, H_2O, N_2 etc…)**

B. Zaniol et al., Rev. Sci. Inst. 91 013103 (2020)

Local plasma parameters: Langmuir probes

- **Local plasma parameters but invasive technique!**
- **Electron branch one must enforce** $a \ln\left[\frac{\pi l}{4a}\right], \lambda_{\mathrm{d}} \ll \inf(\lambda_{\mathrm{e}}, r_{\mathrm{l}})$
- **Often not fullfilled due to RF and B !** *V. Godyak, J. Phys. D.: Appl. Phys 44 233001 (2011)*
- **Solution for correct electron branch evaluation:** *FF. Chen, Plas. Sources Sci. Techn. 21 055013 (2012)*
- **Estimation of the electronegativity** *H. Amemiya, J. Phys. D: Appl. Phys. 23, 999 (1990)* *J. Bredin et al., Phys. Plas. 21 123502 (2014)*

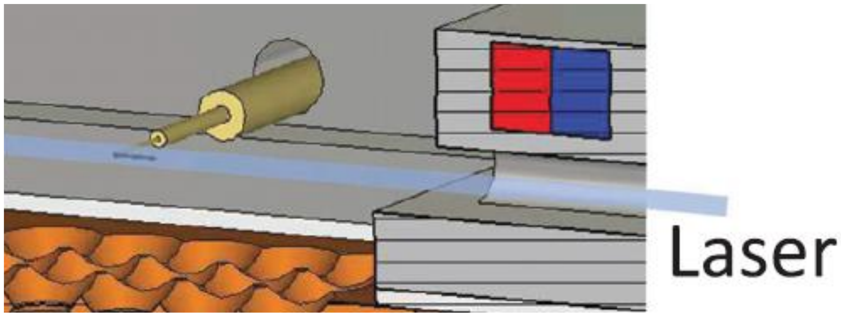
Electron rich **Ion-Ion**



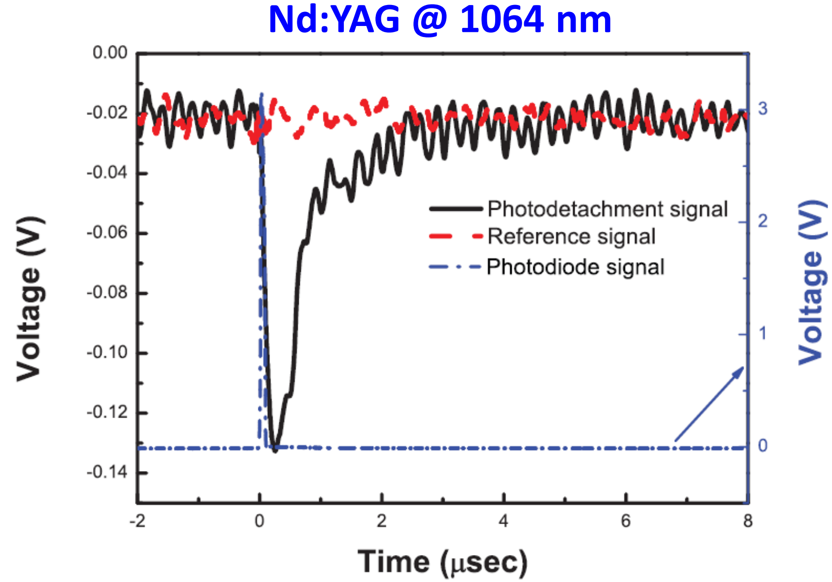
L. Schiesko et al., Plas. Phys. & Contr. Fusion 53 085029 (2011)

Negative ion density (LOS integrated)

1. Laser photodetachement coupled to RF compensated Langmuir probe

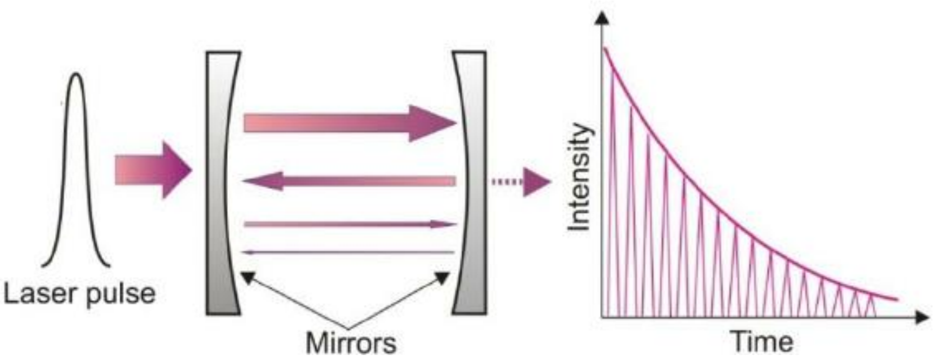


$$\Delta I_{es}/I_{es0} = n_{H^-}/n_e$$

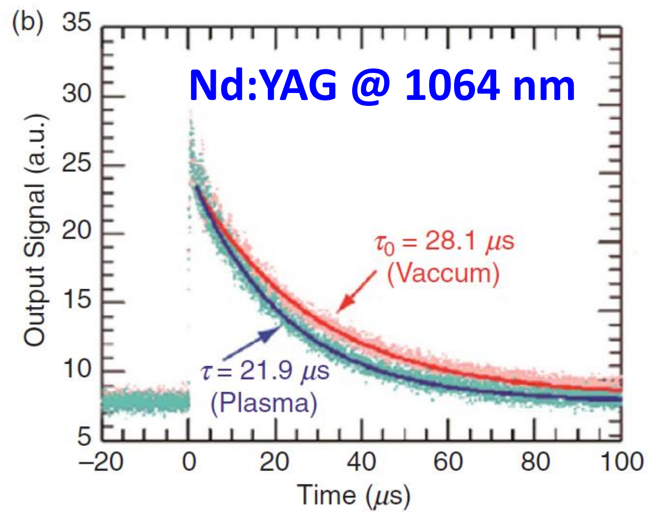


M. Park et al., Phys. Plas. 28 023505 (2021)

2. Cavity ring-down spectroscopy



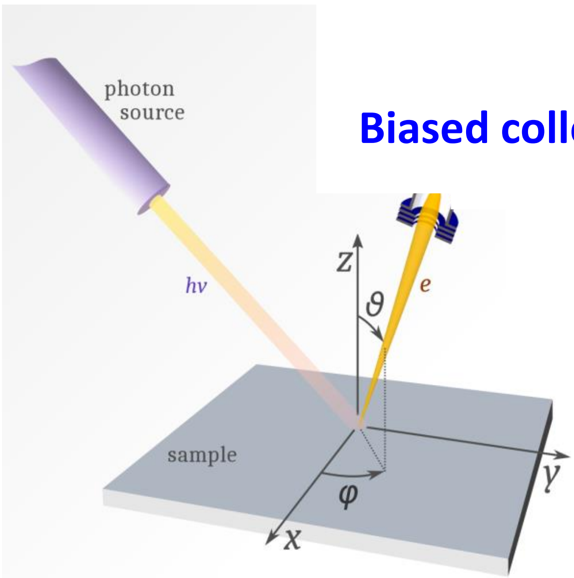
- **Fit of the laser exponential ringdown decay ➡ H^- density**



H. Nakano et al., J. Instr. 11 C03018 (2016)

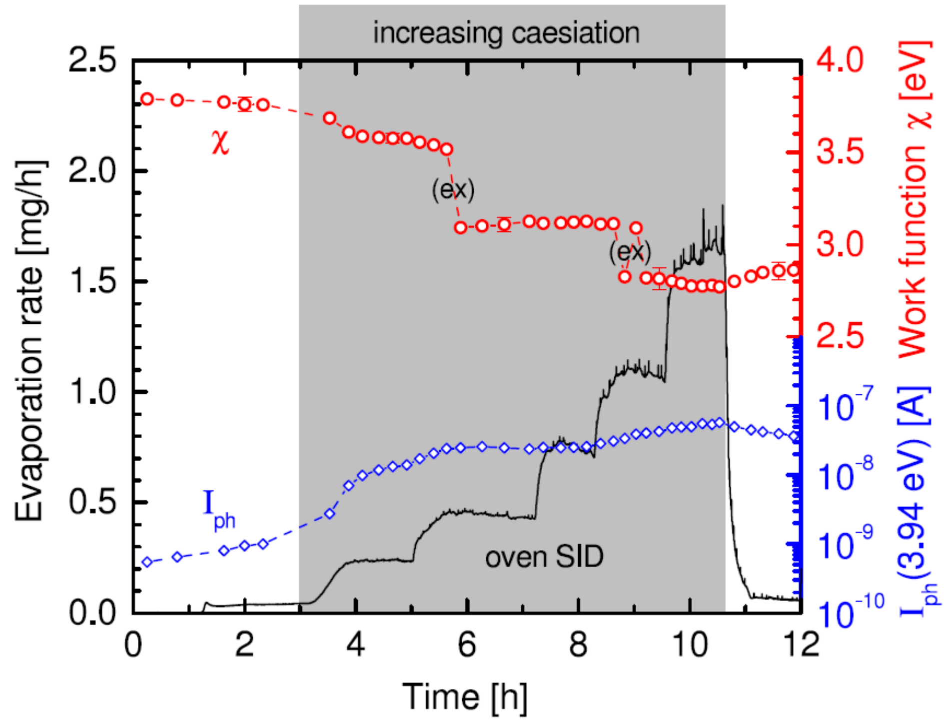
Work function evaluation

Photoelectron emission



Biased collector

$$\ln\left[\frac{I_{\mathrm{ph}}(h\nu)/F(h\nu)}{T^2}\right] = A + \ln\left[f\left(\frac{h\nu}{k_{\mathrm{B}}T} - \frac{\chi}{k_{\mathrm{B}}T}\right)\right]$$



R. Friedl et al., Rev. Sci. Inst. 87 043901 (2016)

- **Photocurrent normalized to the square root of the surface temperatures is prop. to the WF χ (plus a constant A)**

Summary of commonly used diagnostics

Diagnostic	Parameter	Global, Local, LoS int.	Invasive	Remarks
Amperemeter	H^- and e^- currents	Global or local	No	Performance
Optical emission spec.	n_e, Te, nH^0 and more…	LoS	No	Absolute calib. + CR model
Langmuir probe (LP)	Typical plasma params.	Local	Yes	Careful analysis
Laser photodet. + LP	nH^-	Local	Yes	Alignment !
Cavity ringdown	nH^-	LoS	No	Alignment !
Fowler	WF	Local	No	Between plasma pulses

__Diagnostics review__: K. Tsumori et al., App. Phys. Rev. 8 021314 (2021)

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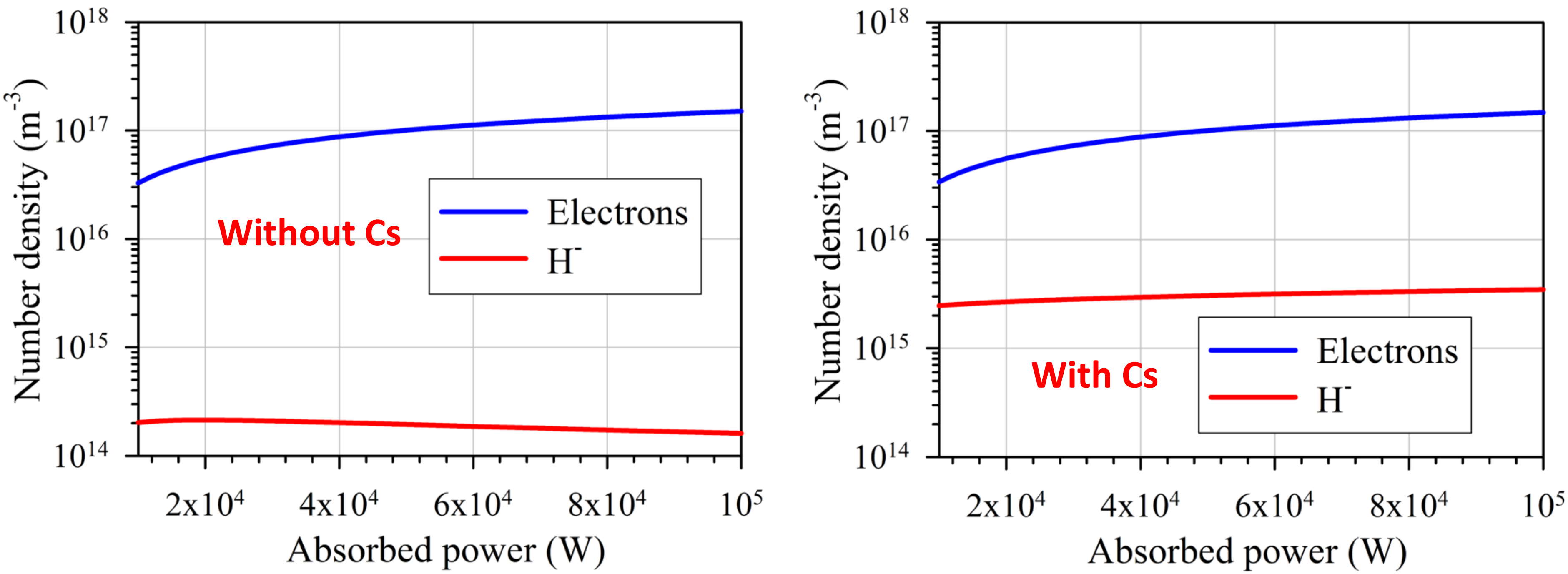
III. **Theory and modelling**
IV. Conclusion

Global model (0D)

➢ Set of coupled particle and power balance equations, neglects spatial derivatives but simple implementation and efficient !

➢ Difficulty: accurate cross sections / rates coefficients needed

LXCat database: E. Carbone et al., Atoms 9, 16 (2021)
NIST database: V. K. Shen et al., NIST Standard Ref. Database 173 http://doi.org/10.18434/T4M88Q
S. H. Scarlett et al., Plasma Sources Sci. Technol. 28, 025004 (2019)



Averkin S. N. et al., AIP Conf. Proc. 2011 050026 (2018)

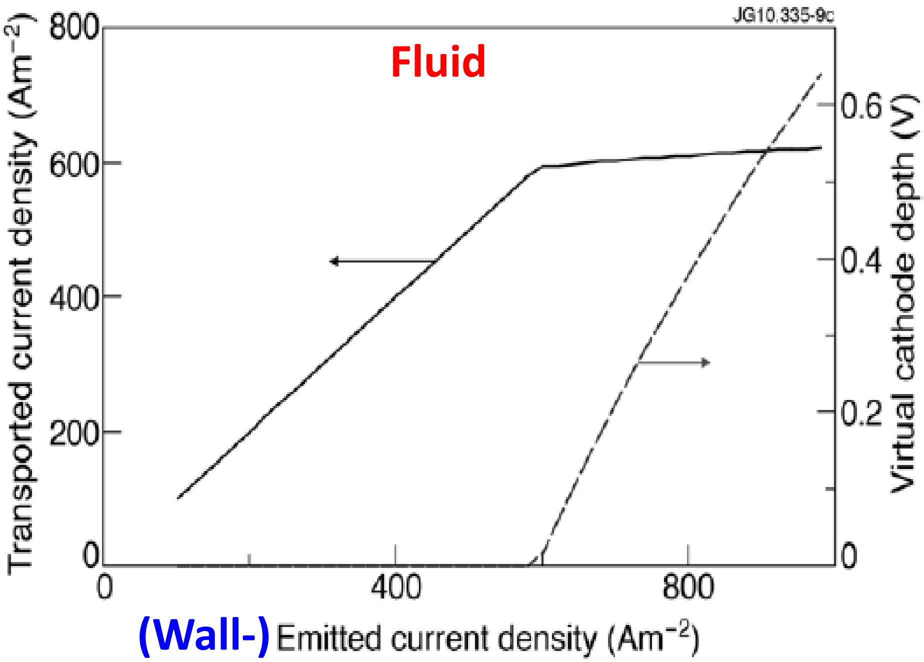
➢ Accurate qualitative results (0D) to determine trends

Review Global Modelling: A. Hurlbatt et al., Plasma Process Polym. 14 1600138 (2017)

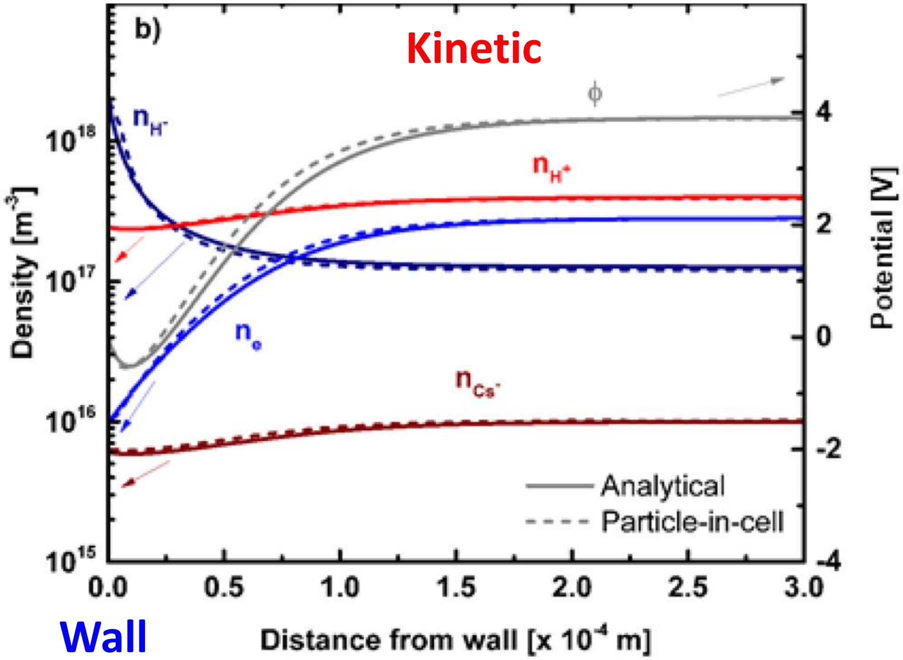
1D fluid and kinetic models

- **Fluid: moments of Boltzmann equation**
- **Kinetic: truncated Maxwellian solutions of the Vlasov eq.**

For plasma-wall interaction



R. McAdams et al., Plasma Sources Sci. Technol. 20, 035023 (2011)

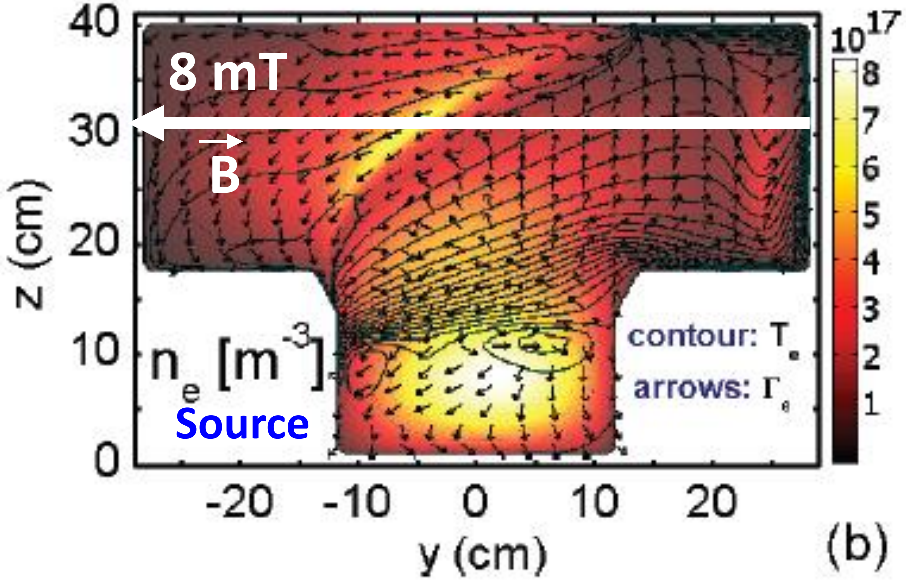


L. Schiesko et al., J. Appl. Phys. 127 033302 (2020)

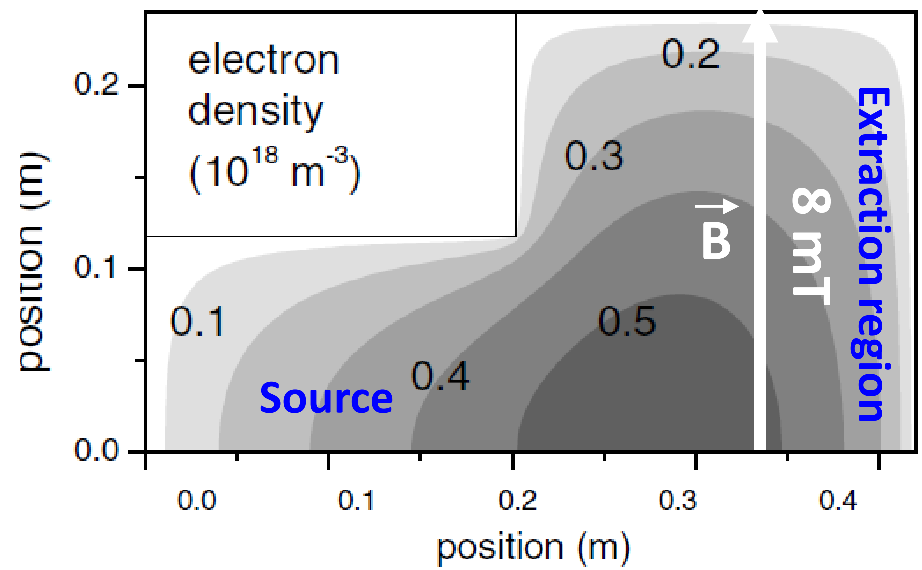
- **Transported negative ions: space-charge limitation (Child-Langmuir)**
- **Very quick and accurate results, useful for parameters scans**

2D fluid for magnetized plasma transport

- **Fluid: moments of Boltzmann equations**
- **Magnetic fields: drifts, at least 2D needed !**



S. Lishev, L. Schiesko et al., AIP Conf. Proc. 1655 040010 (2015)

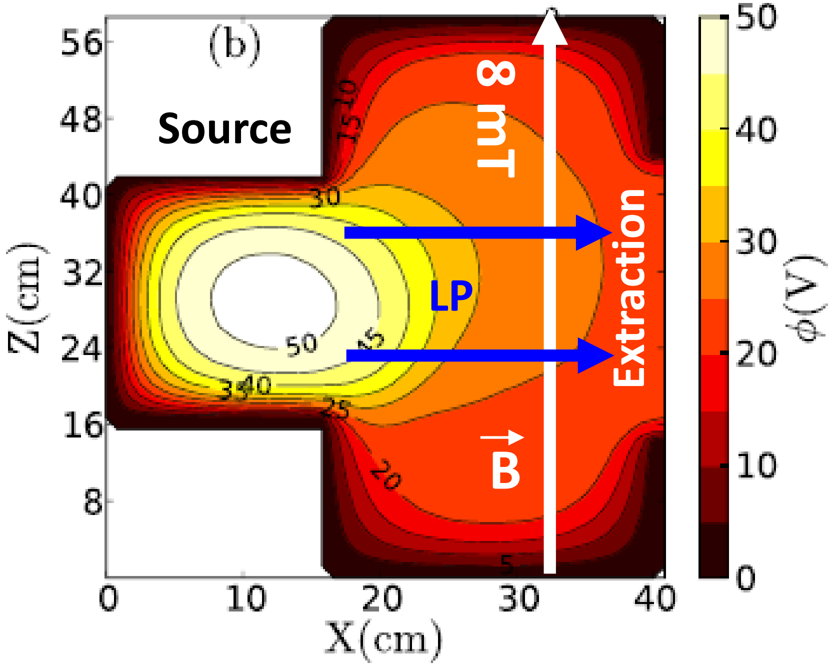


G. J. M. Hagelaar et al., Plasma Sources Sci. Technol. 20, 015001 (2011)

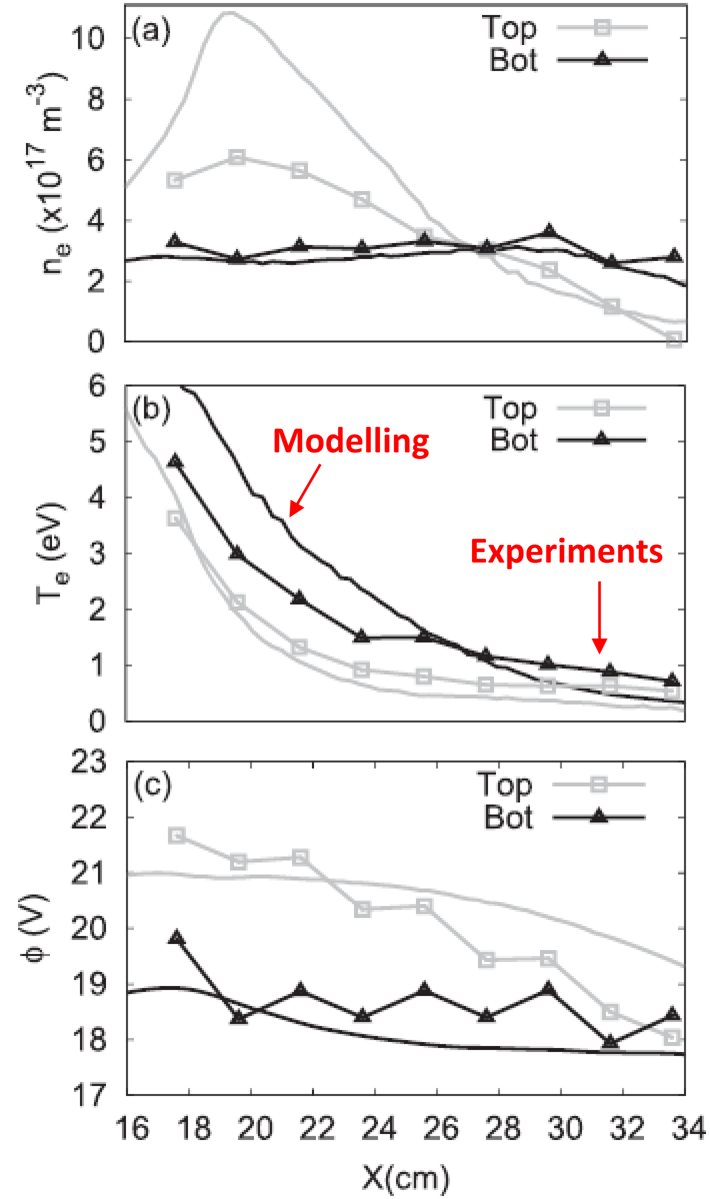
- **Very general models including neutrals (Navier-Stockes eqs.) and surface processes as well as self-consistent RF coupling (Maxwell eqs.)**
- **Reproduces non-trivial charged particles transport: $\vec{E}\times\vec{B}$, diamagnetic drifts**

3D Particle-In-Cell (PIC): bulk plasma

- **PIC: self-consistent particle method**
- **3D: realistic transport w/r magnetic field**
- **Requires massive parallelisation (OpenMP, OpenMPI, OpenAcc etc…)**



G. Fubiani et al., Phys. Plas. 20 113511 (2013)

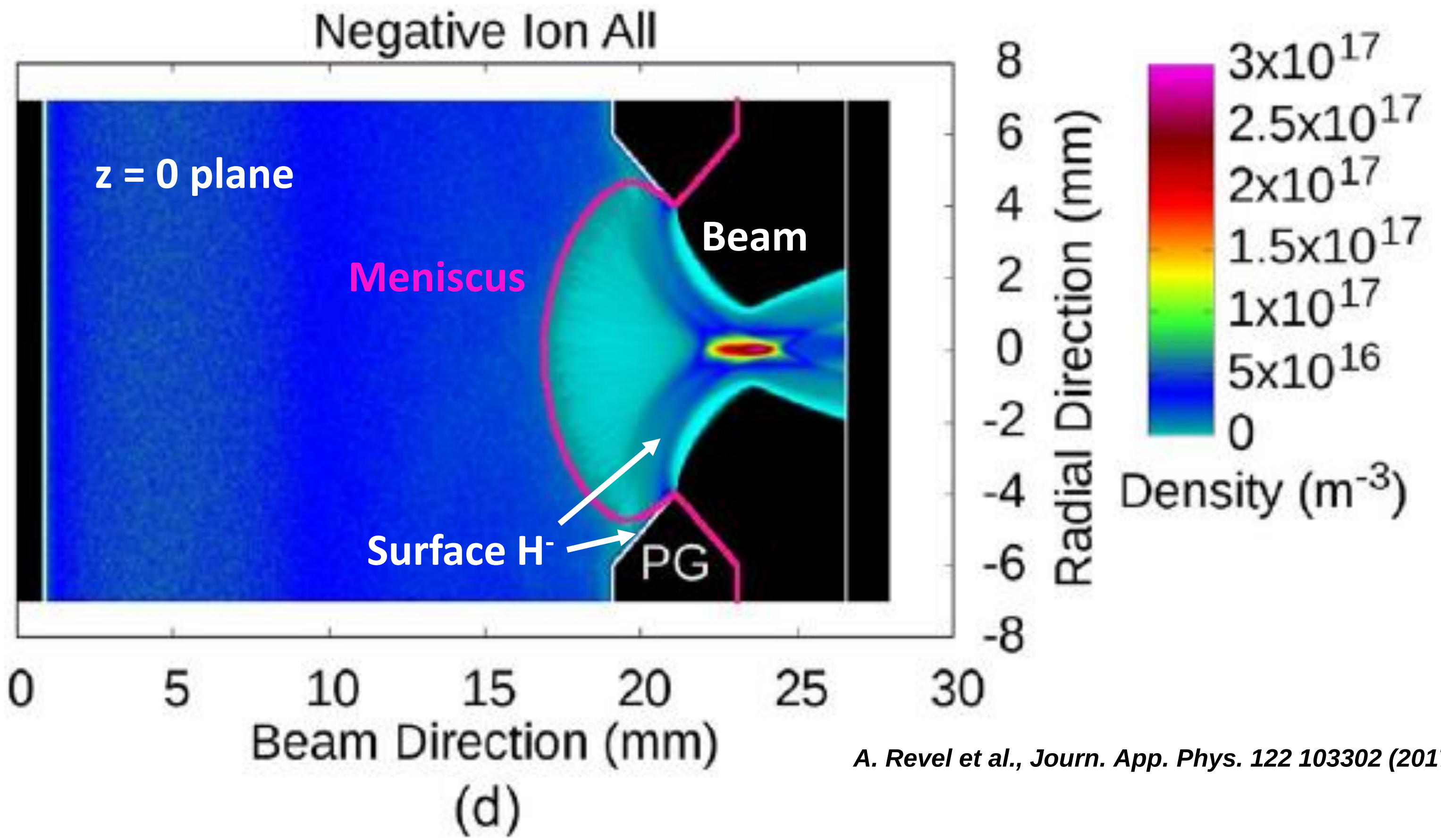


- **Very good agreement with Langmuir probe measurements**
- **3D: simulations close to reality !**

Modelling: G. Fubiani et al., New J. Phys. 19 015002 (2017)
Experimental: L. Schiesko et al., Plas. Phys. Cont. Fus. 54 105002 (2011)

3D Particle-In-Cell (PIC): H^- extraction

- **Understanding the negative ion extraction is a key parameter**
- **2D cannot fully unveil transport and extraction of H^- in a magnetized plasma**



A. Revel et al., Journ. App. Phys. 122 103302 (2017)

- **Discrimination between direct and indirect extraction of surface generated H^-**
- **Beam formation and properties such as divergence, perveance …**

Summary theory and modelling

- Different possibilities for modelling
  - qualitative, qantitative, predictive…
- Global model / 0D
  - volume averaged results, fast implementation,
  - limitations on known rate coefficients is the major drawback
- 1D kinetic / fluid / PIC
  - usually for plasma-wall interaction, quick and very accurate results
- 2D fluid or PIC
  - transport of magnetized plasma species, presence of drifts
- 3D PIC
  - simulations as close as possible to reality needed,
  - negative ion generation, transport and extraction,
  - time consuming, small volume simulated, cluster needed

Outline

I. Work principle
   i. Volume negative ions generation and destruction mechanisms, example of source
   ii. Surface negative ions generation, example of source
II. Diagnostics
III. Theory and modelling
IV. **Conclusion**

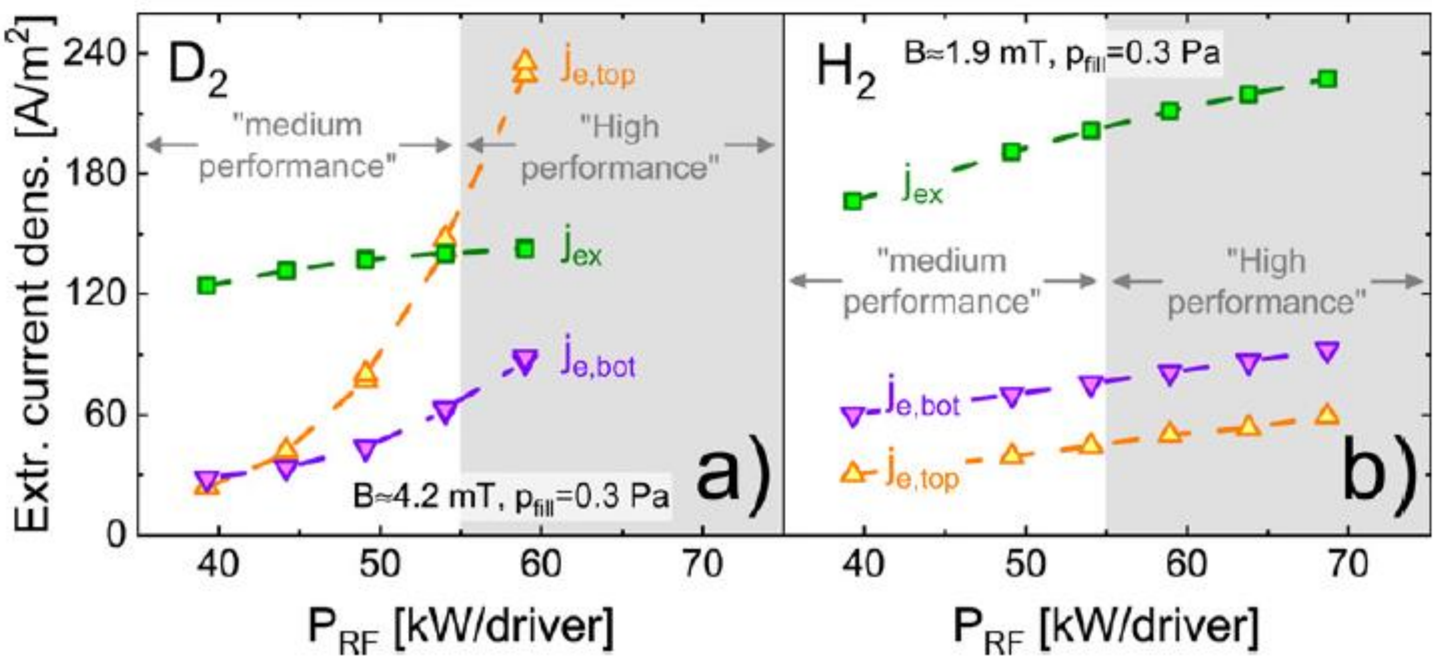
Conclusion

- Negative ion sources are an unavoidable setup for high energy accelerators / neutral beam injectors
- Main generation and destruction mechanisms well understood:
    - Volume: dissociative attachement to rovibrationnaly excited H_2
    - Surface: backscattering or sputtering of an adsorbed hydrogen as a negative ion, low WF material, Cs evaporation or Cs-free
- Different plasma sources available: RF, ECR, Filament…
- For both generation mechanisms, > 10 eV and < 2eV are needed !
- Use of a magnetic field: separation of the source in a high Te and a low Te
- Magnetic field very effective, but generates drifts !
- Many diagnostics can be used for plasma parameters, Cs monitoring, densities…
- Modelling and theory well developped: 0D, 1D, 2D and 3D models /particle-in-cell give hints on the underlying physics and can be predictive

Challenges: isotopic effect

Hydrogen vs. Deuterium

Review Deuterium operation: *M. Bacal et al., Plasma Sources Sci. Technol. 29, 033001 (2020)*



Short pulses

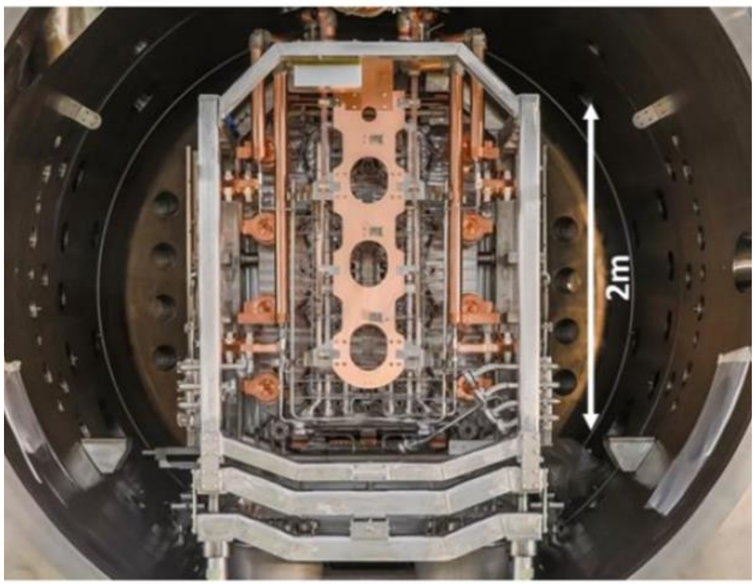
D. Wünderlich et al., Nucl. Fusion 61 096023 (2021)

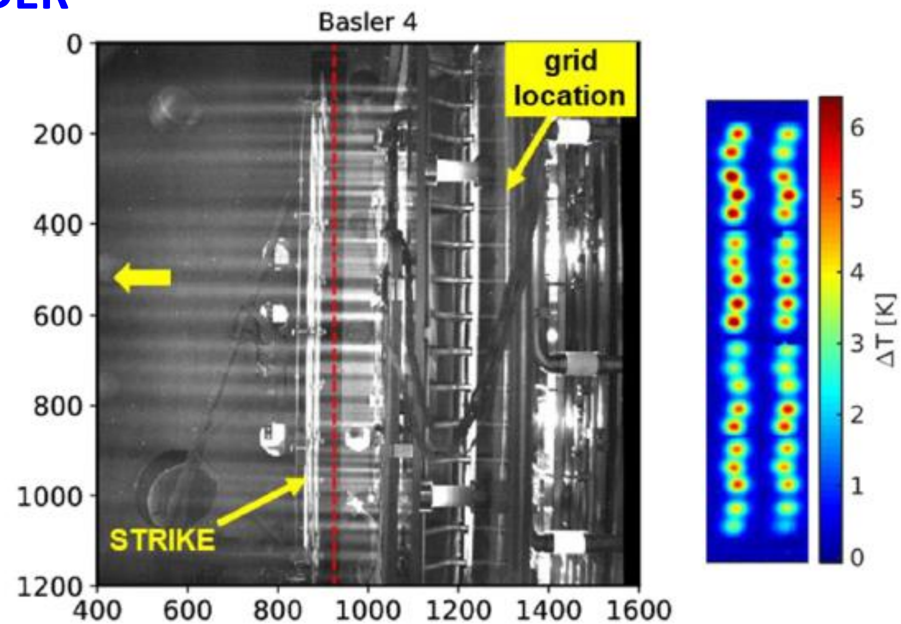
- **Extracted D^- < H^- and more co-ext. electrons in D_2 although stronger $\vec{B}$**
- **Mitigation possible**
  - **with the use of potential rods near the extraction system**
  - **'Cs shower': oven evaporating near the first accelerator grid**

Challenges: ITER NBI

- ITER requirements: 40 A D^- at 1 MeV during 1h cw, never simultaneously achieved !
- At Consorzio RFX, SPIDER: full-scale prototype (100 kV), MITICA: entire ITER NBI (1 MV) under construction

SPIDER





G. Serianni et al., Rev. Sci. Inst. 91 023510 (2020)

- Beam uniformity, div < 7 mrad, Cs uniformity, 1 MV voltage holding, co-ext. electrons, thermal and thermomechanical issues, radiations from ITER …