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ISIS Neutron and
Muon Source

The role of plasma-facing surfaces in high-current negative hydrogen ion sources

Olli Tarvainen, Dan Faircloth, Erin L. Flannigan

STFC ISIS Pulsed Spallation Neutron and Muon Source

Robert F. Welton

Spallation Neutron Source, Oak Ridge National Laboratory



Introduction

This is a review talk

The focus is on the importance of

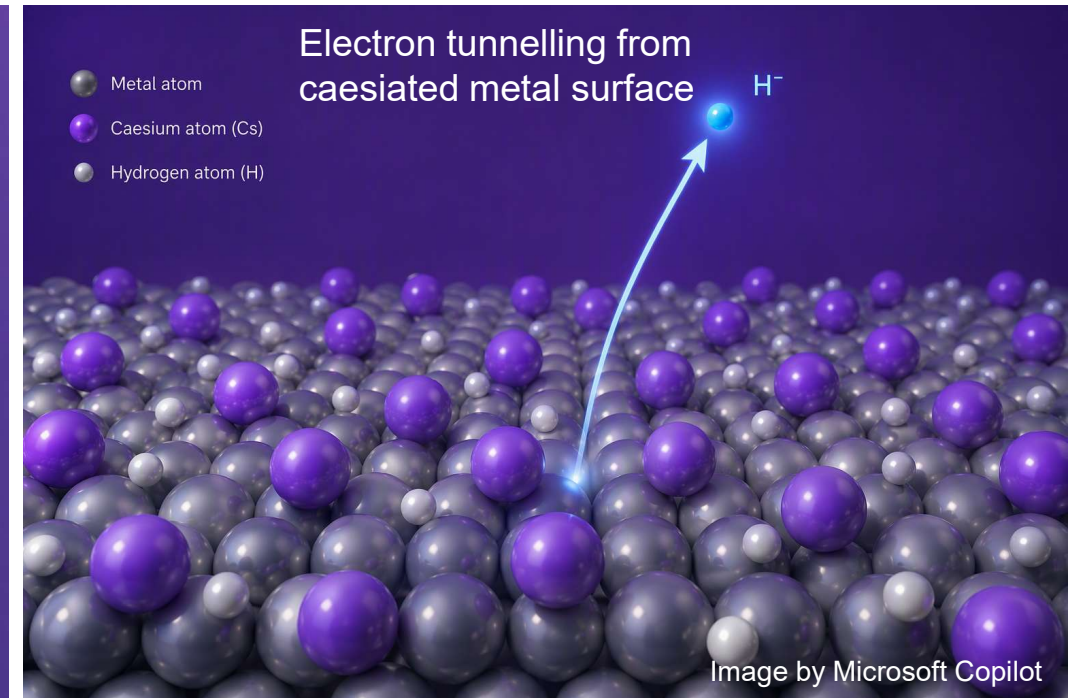
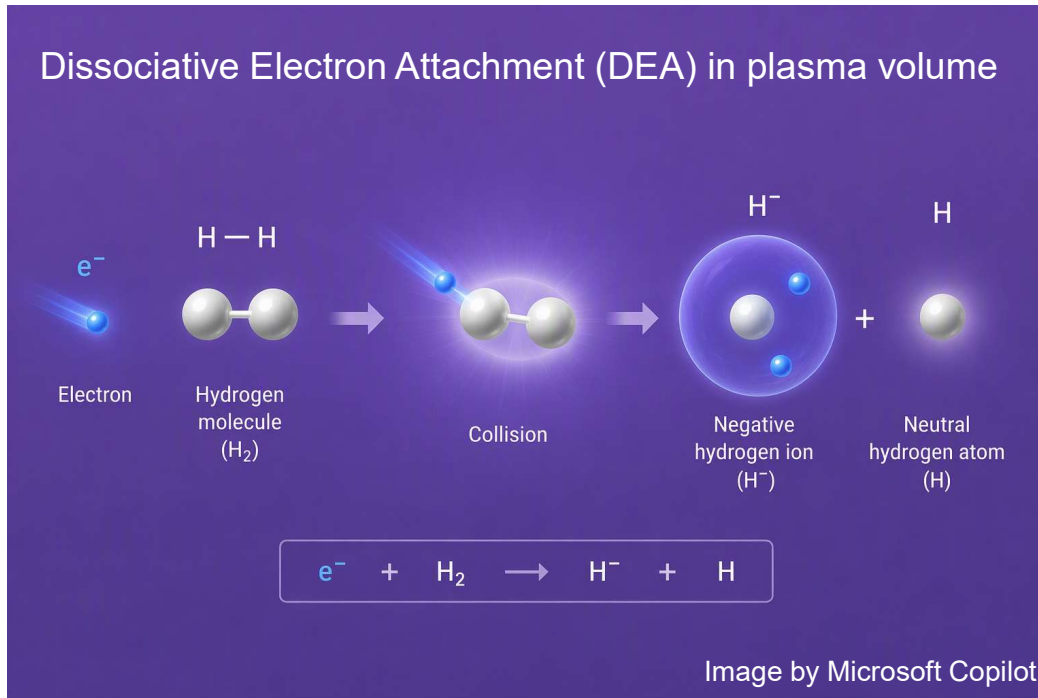
- Discharge power coupling method
- Plasma-facing material selection

for optimising the H^- (or D^-) yield especially in the surface production mode.

Predictions on the relative H^- yields from different caesiumated metal surfaces.

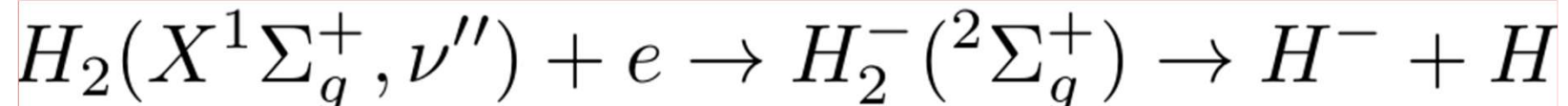
Recommendations for experimental and theoretical work.

Two main production channels of H^-

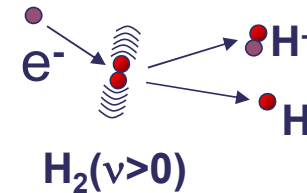
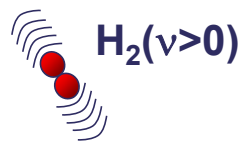
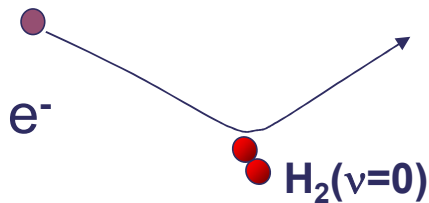


How does the discharge power coupling and surface material selection affect each?

Dissociative Electron Attachment (DEA)



Requires ro-vibrational excitation of the H₂ molecules and high (cold) electron density



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PHYSICAL REVIEW LETTERS

4 JUNE 1979

H⁻ and D⁻ Production in Plasmas

M. Bacal and G. W. Hamilton^(a)

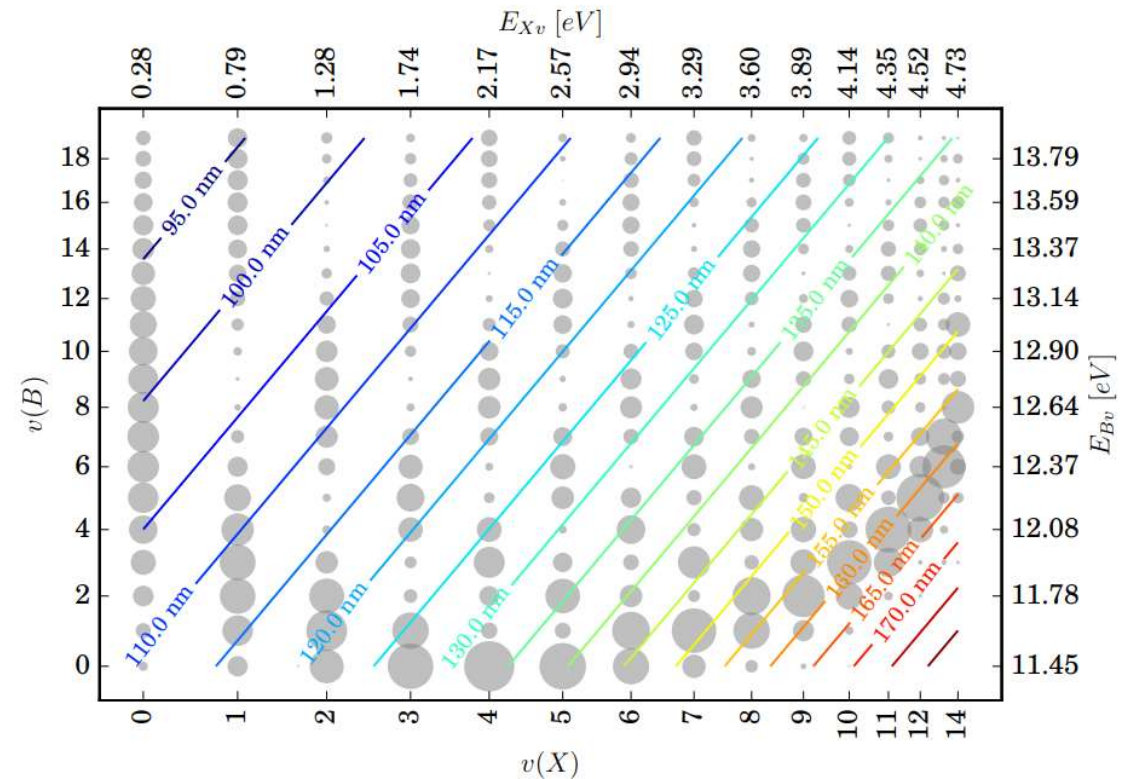
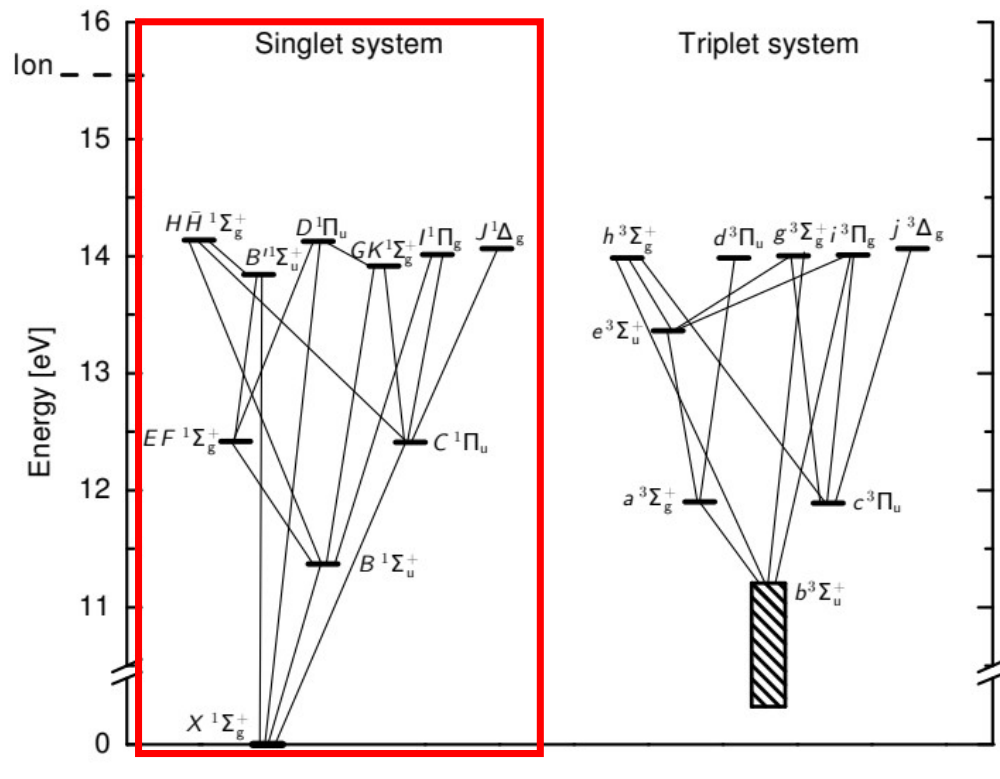
Laboratoire de Physique des Milieux Ionisés, Groupe de Recherche du Centre National de la Recherche Scientifique, Ecole Polytechnique, 91128 Palaiseau Cedex, France

(Received 2 January 1979)

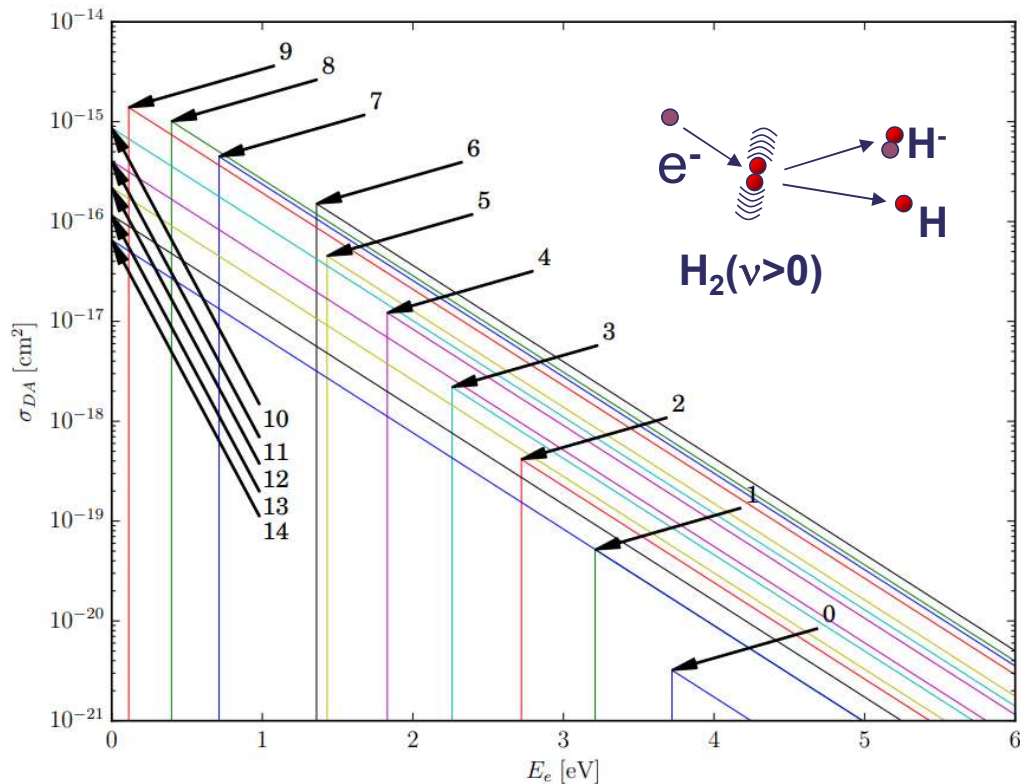
H₂ vibrational excitation

The predominant channel for the excitations of high vibrational levels in the plasma volume is electronic excitation to B¹Σ_u⁺ and C¹Π_u singlet states, followed by VUV-emitting radiative decay to high vibrational levels (v'' ≥ 5) of the X¹Σ_g⁺ ground state

- J.R. Hiskes, J. Appl. Phys. 51 4592, (1980) -



Dissociative Electron Attachment (DEA)



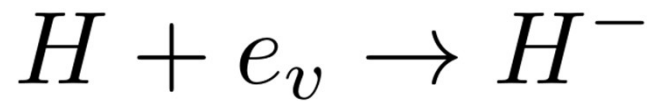
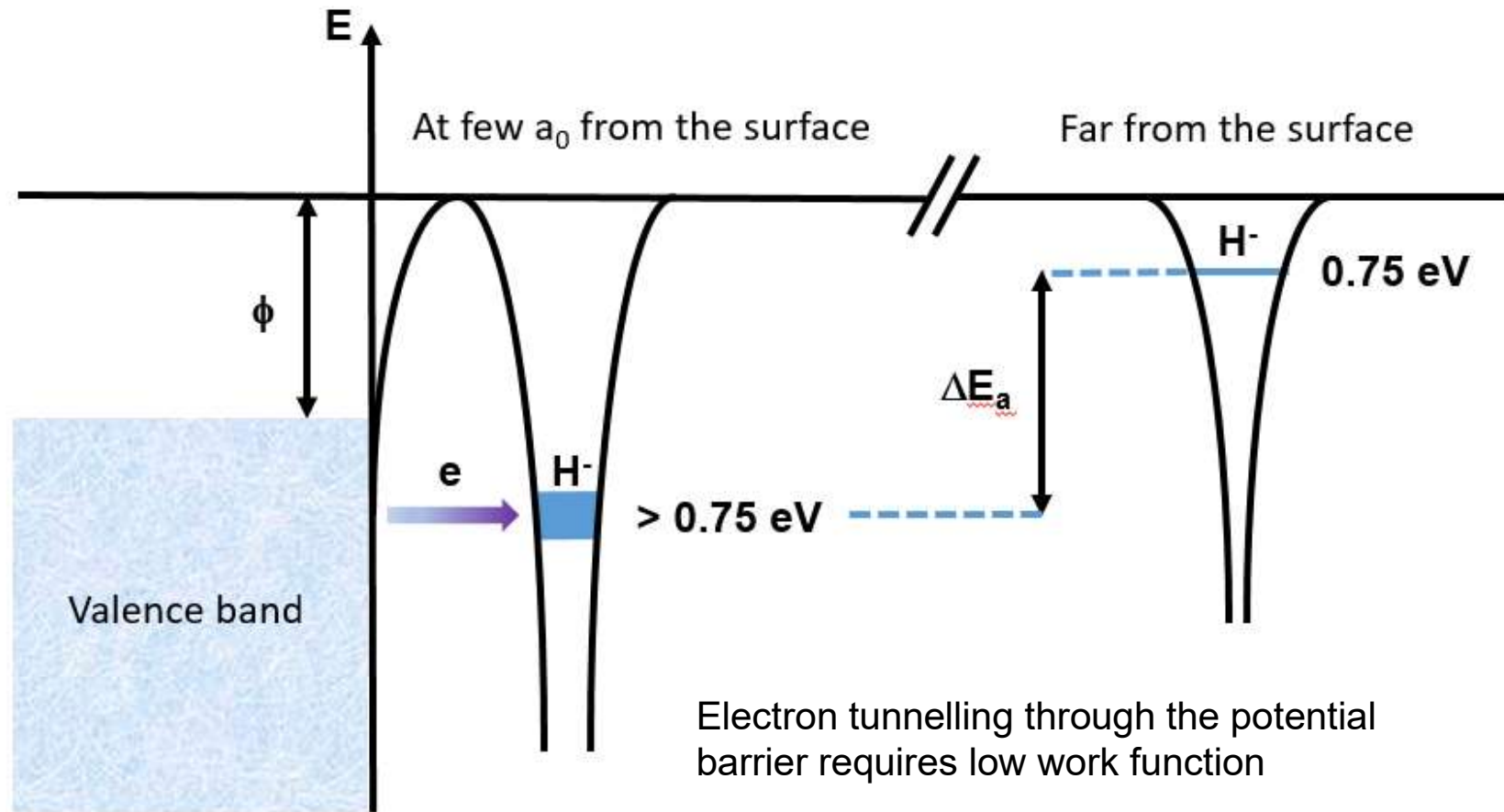
The DEA cross section increases by 6 orders of magnitude with increasing vibrational state.

At the same time the threshold DEA electrons energy becomes very low.

Vibrationally excited molecules and low energy electrons!

Either one can be a bottle neck limiting the DEA rate...

Surface production of H^-

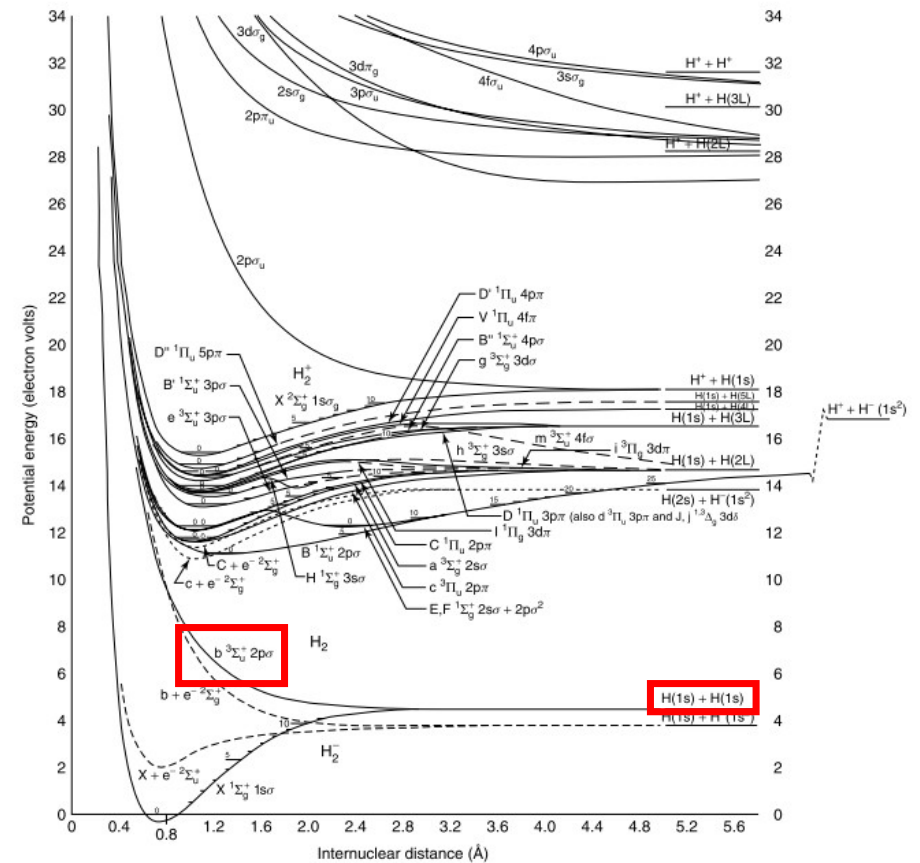
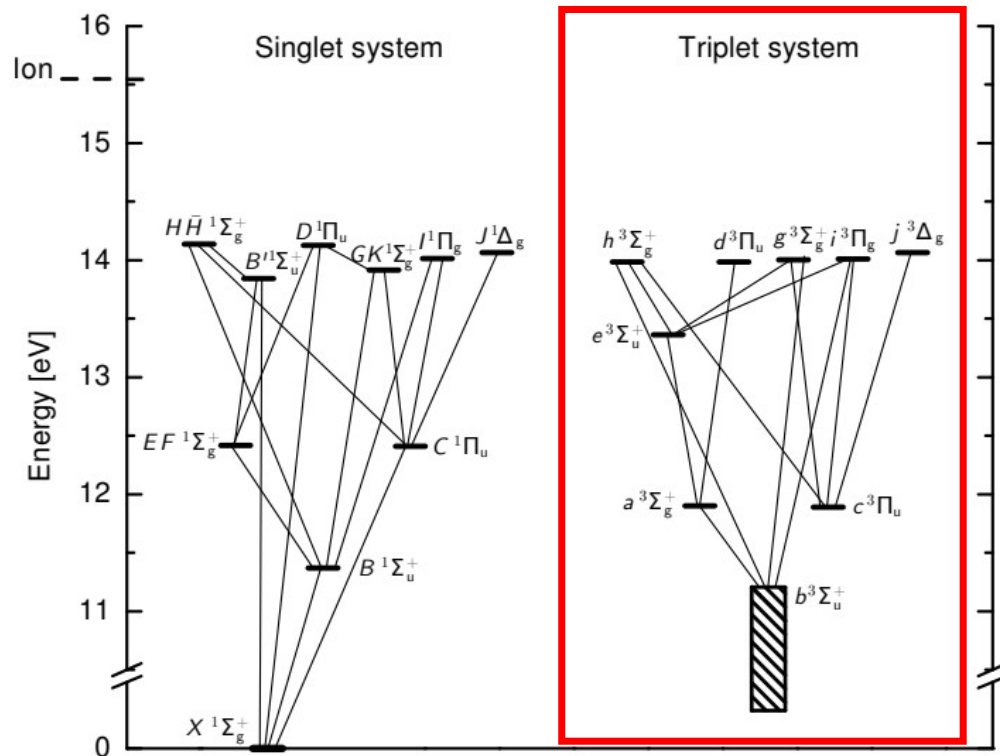


H₂ dissociation

The most significant dissociation channel of the hydrogen molecule is excitation to triplet states.

The majority of triplet excitations lead to the repulsive $b^3\Sigma_u^+$ state.

- R. Celiberto *et al.* At. Data Nucl. Data Tables, 77(2):161, 2001 –



Optimising volume vs. surface production

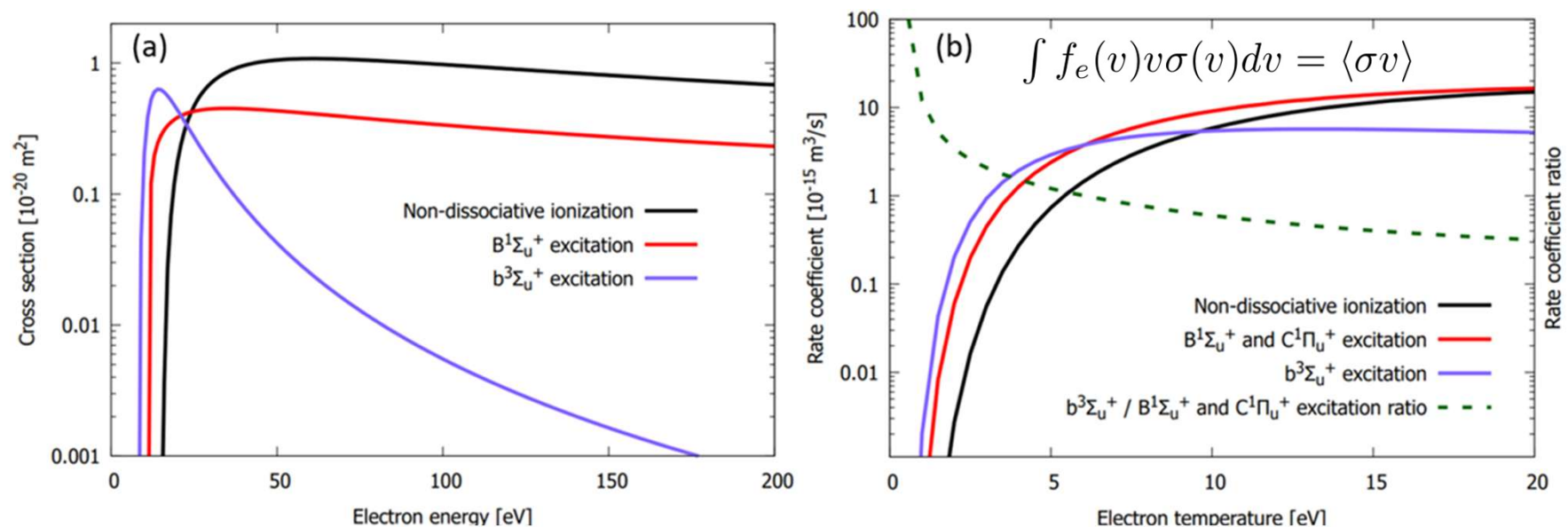
Optimising the volume production requires optimising the vibrational excitation of the H_2 molecules.

This can be achieved by optimising electronic excitation to singlet states.

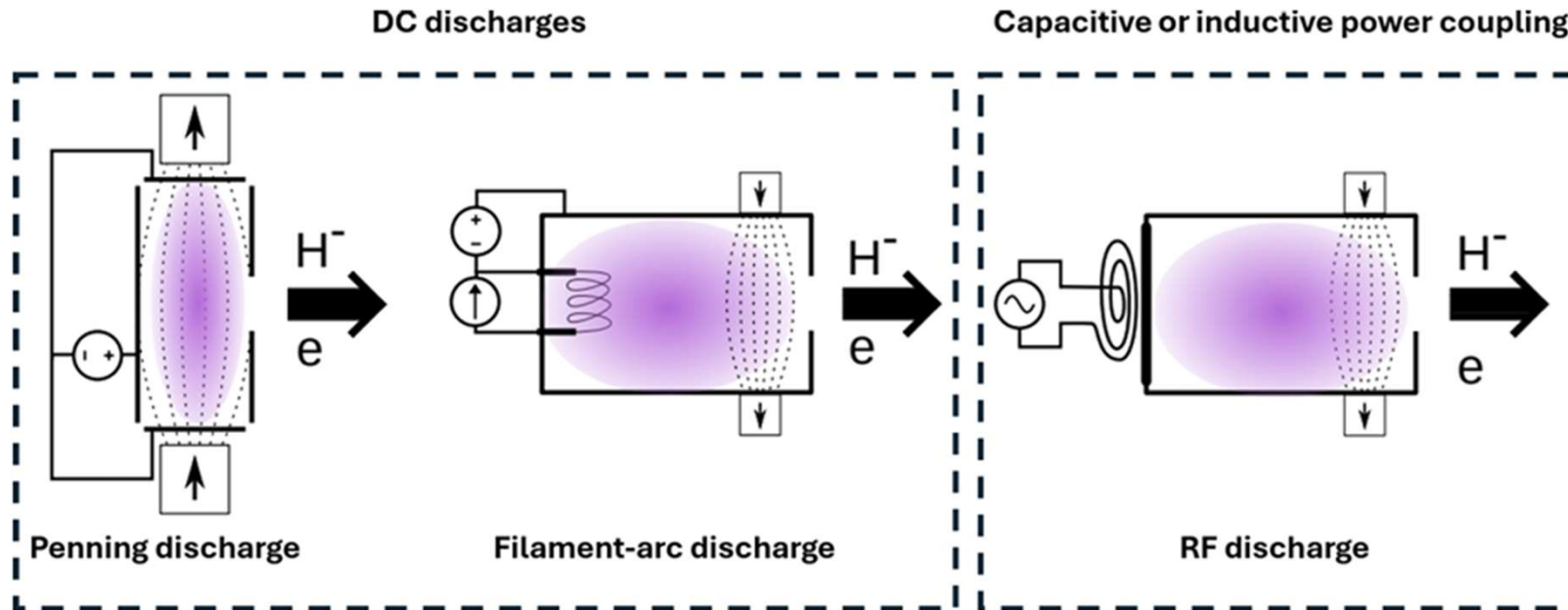
Optimising the surface production requires optimising molecular dissociation to supply H atoms.

This can be achieved by optimising electronic excitation to triplet states.

The most probable excitations to singlet and triplet states are $X^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$ and $X^1\Sigma_g^+ \rightarrow b^3\Sigma_u^+$
Their cross sections peak at very different electron energy affecting the rate coefficient ratio vs T_e



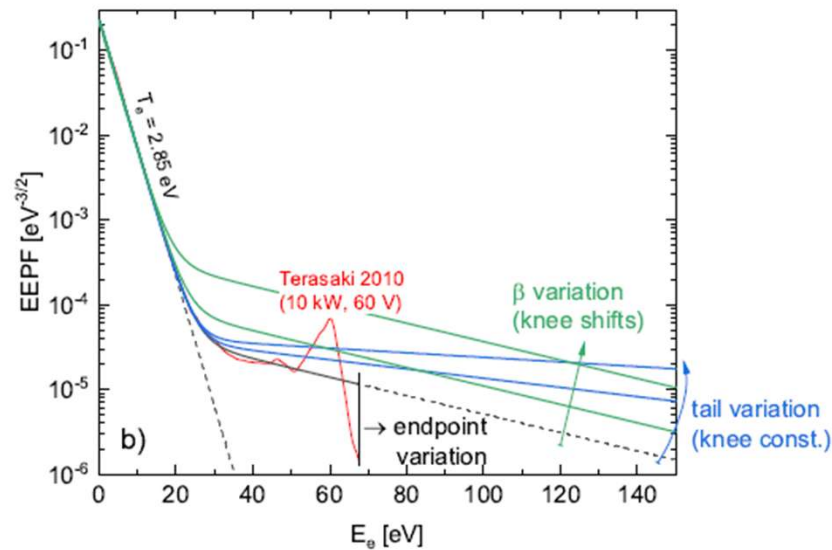
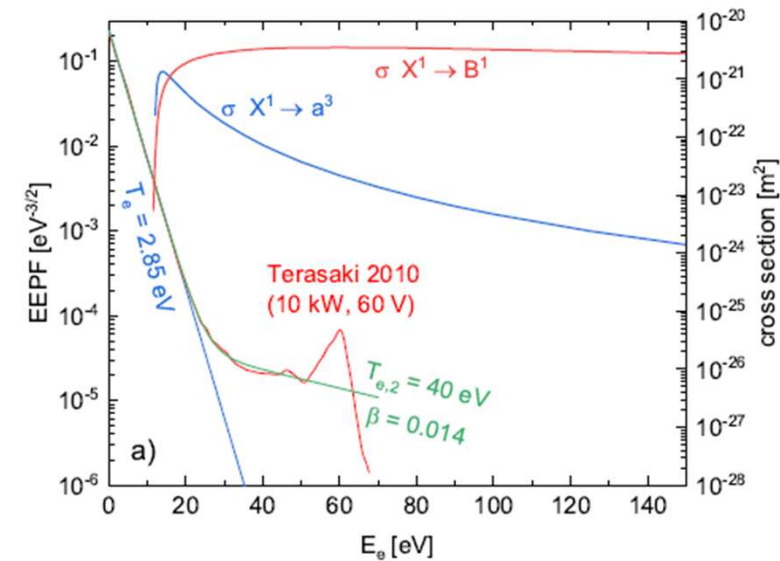
Importance of discharge type



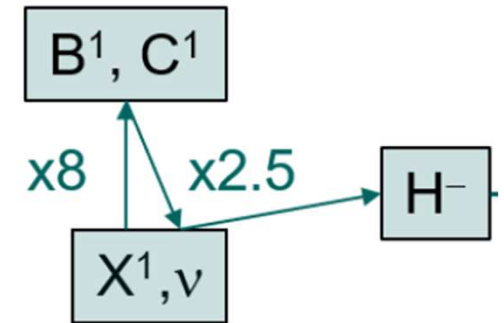
Electron Energy Distribution (EED)
with hot tail favours volume production

Maxwellian EED better
suited for surface production

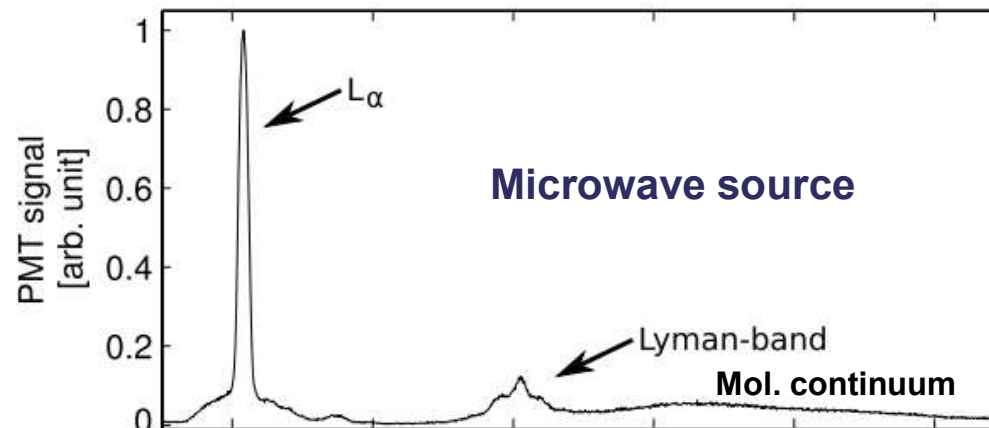
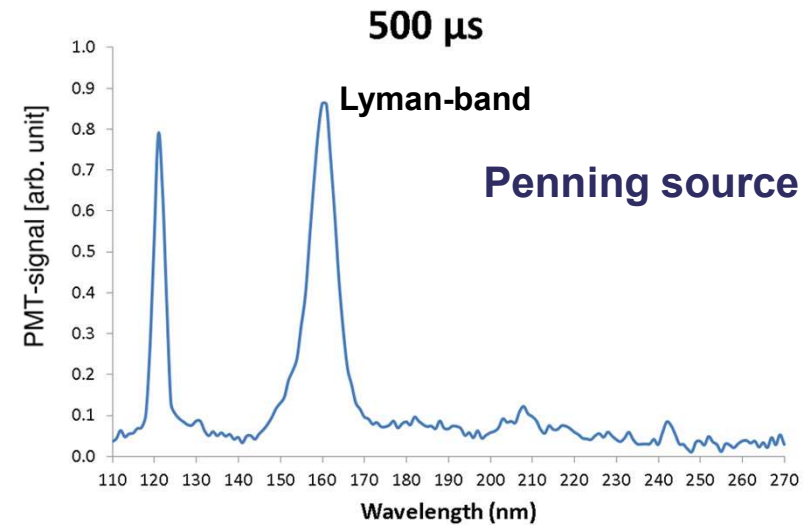
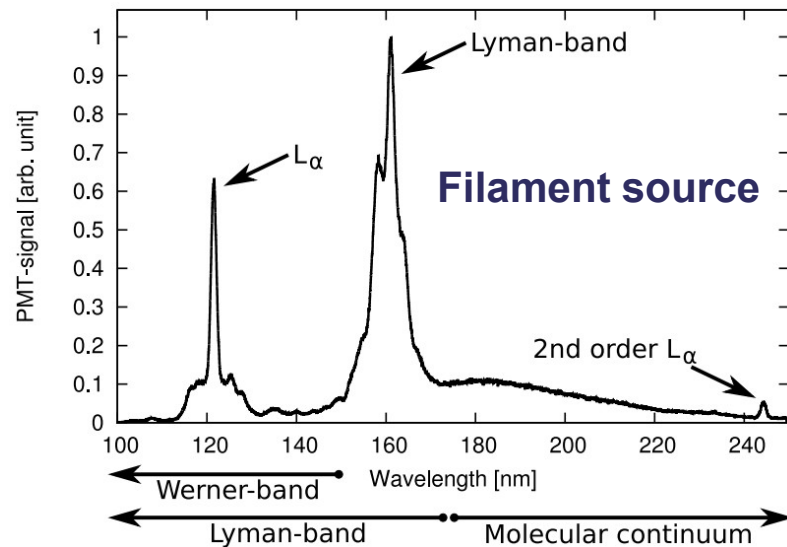
Collisional radiative modelling



Hot tail vs. Maxwellian



VUV-spectroscopy

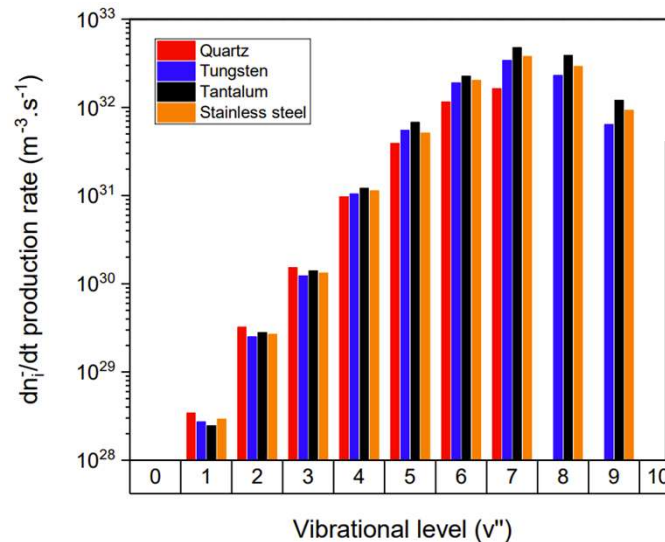
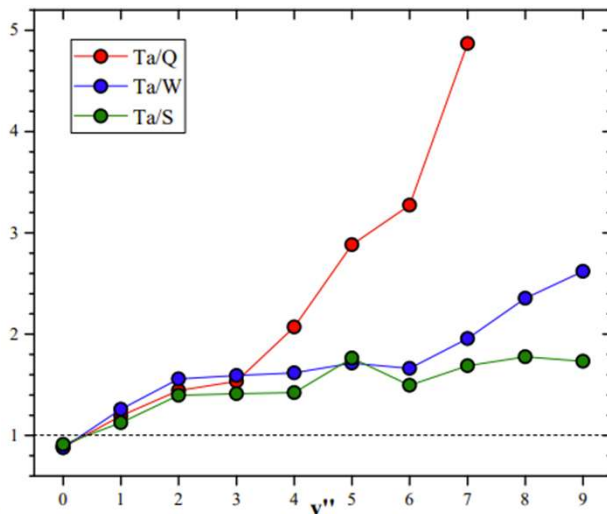


The discharge type matters!

Surfaces affecting vibrational excitation

The vibrational distribution of H_2 molecules can be affected by surface interactions

- Recombination of atoms
- Auger relaxation of the repulsive $b_3\Sigma_u^+$ state quenching to high vibrational level
- Electronic recombination and dissociation of H_3 compound molecule



Journal of Quantitative Spectroscopy and
Radiative Transfer

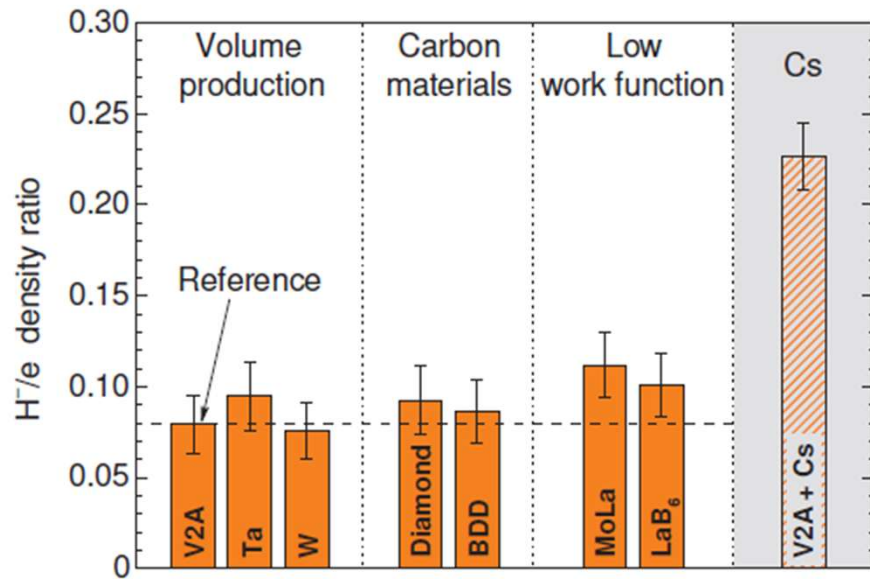
Volume 257, December 2020, 107325



Direct measurements of electronic ground state ro-vibrationally excited D_2 molecules produced on ECR plasma-facing materials by means of VUV-FT absorption spectroscopy

S. Béchu^{1 a}, J.L. Lemaire^{2 b}, L. Gavilan^c, S. Aleiferis^{d e f}, V. Shakhmatov^g,
Yu.A. Lebedev^h, D. Fombaronⁱ, L. Bonny^j, J. Menu^j, A. Bès^j, P. Svarnas^j, N. de
Oliveira^k

Does this translate to H⁻ density?



Microwave source with electrostatic filter

RESEARCH ARTICLE | AUGUST 09 2017

Efficiency of Cs-free materials for negative ion production in H₂ and D₂ plasmas

R. Friedl; U. Kurutz; U. Fantz

+ Author & Article Information

AIP Conf. Proc. 1869, 030022 (2017)

<https://doi.org/10.1063/1.4995742>

VUV-irradiance measurement (microwave source)

Table 3. Ratios of rate coefficients (plasma process versus Lyman-band or molecular continuum emission) and volumetric reaction rates in the line of sight volume.

	Ratio of rate coefficients	Volumetric rate [cm ⁻³ s ⁻¹]
Lyman-band		
Molecule ionization	0.1–1.3	0.14–2.1 × 10 ¹⁶
Excitation ($B^1\Sigma_u^+$, $C^1\Pi_u$)	1.4–1.8	1.8–2.8 × 10 ¹⁶
Excitation ($\nu \geq 5$) ($B^1\Sigma_u^+$, $C^1\Pi_u$)	0.8–1.1	1.1–1.7 × 10 ¹⁶
Dissociation ($B^1\Sigma_u^+$, $C^1\Pi_u$)	0.2–0.3	0.3–0.4 × 10 ¹⁶
Molecular continuum		
Dissociation (via $b^3\Sigma_u^+$)	2.2–8.5	2.8–12 × 10 ¹⁶
Excitation $c^3\Pi_u$ state, max	0.6–1.8	0.8–2.5 × 10 ¹⁶

Each molecule is ionised, excited and dissociated several times as it passes through the ion source

PAPER

VUV irradiance measurement of a 2.45 GHz microwave-driven hydrogen discharge

J Komppula, O Tarvainen, T Kalvas, H Koivisto, R Kronholm, J Laulainen and P Myllyperkiö

Published 13 August 2015 • © 2015 IOP Publishing Ltd

[Journal of Physics D: Applied Physics](#), Volume 48, Number 36

Citation J Komppula et al 2015 *J. Phys. D: Appl. Phys.* **48** 365201

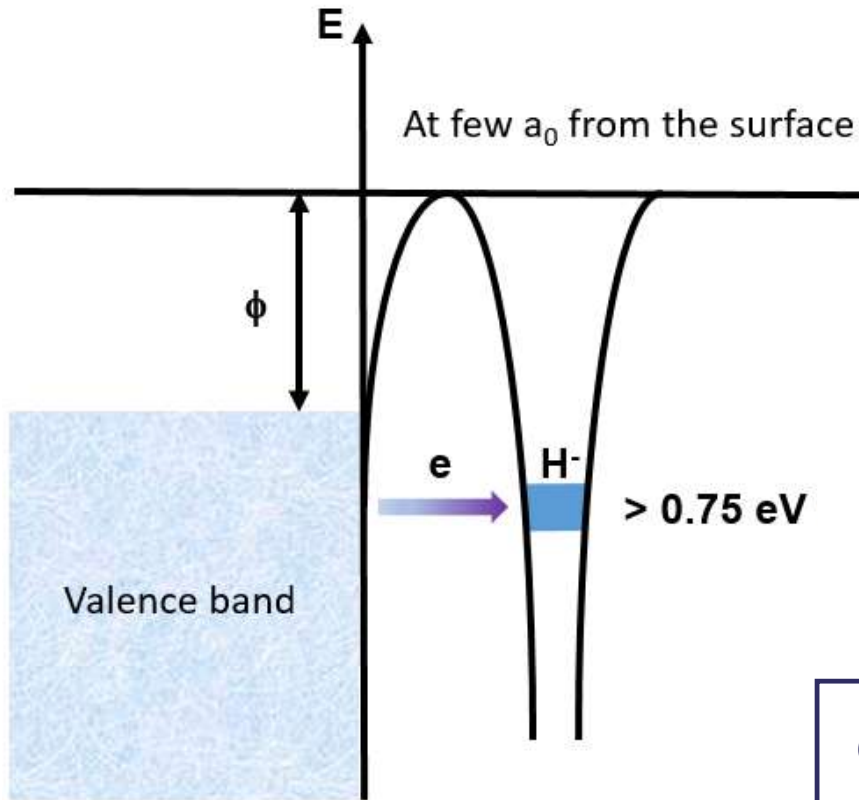
DOI 10.1088/0022-3727/48/36/365201

The surfaces contribute to volume production of H^- via DEA if the vibrational excitation rate via volumetric processes is not sufficient.

In the opposite case the cold electron density is the limiting factor.

Altogether it can be expected that the contribution of surfaces on DEA depends on the discharge type and filter field strength.

Surface production of H⁻



Electron transition rate

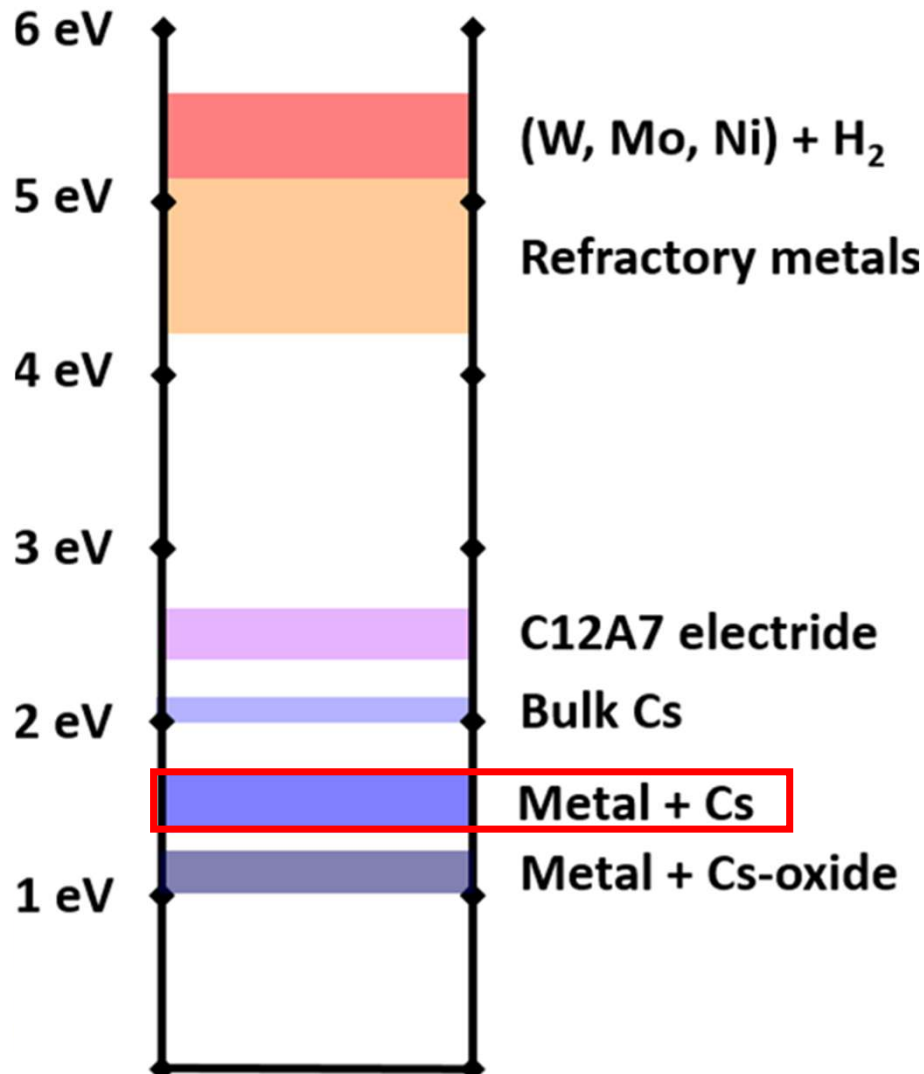
$$R = 2\pi \Sigma |T_i|^2 n_e$$

H⁻ survival probability

$$p_- \simeq \frac{2}{\pi} e^{-\frac{\pi(\phi - E_a)}{2av_z}}$$

Conduction electron density and surface work function

Work function



$$\Delta\phi_m \simeq -1.24\left[\phi_0 - \frac{1}{2}(I_A + E_A)\right]$$

$$\phi_{\min} = \phi_0 + \Delta\phi_m \simeq 2.707 - 0.24\phi_0$$

$$\phi(\theta) = \phi_0 + \frac{6\Delta\phi_m}{(3 - \theta_m)\theta_m}\theta - \frac{3\Delta\phi_m(\theta_m + 1)}{(3 - \theta_m)\theta_m^2}\theta^2 + \frac{2\Delta\phi_m}{(3 - \theta_m)\theta_m^2}\theta^3$$



Surface Science
Volume 175, Issue 1, 1 September 1986, Pages 226-240



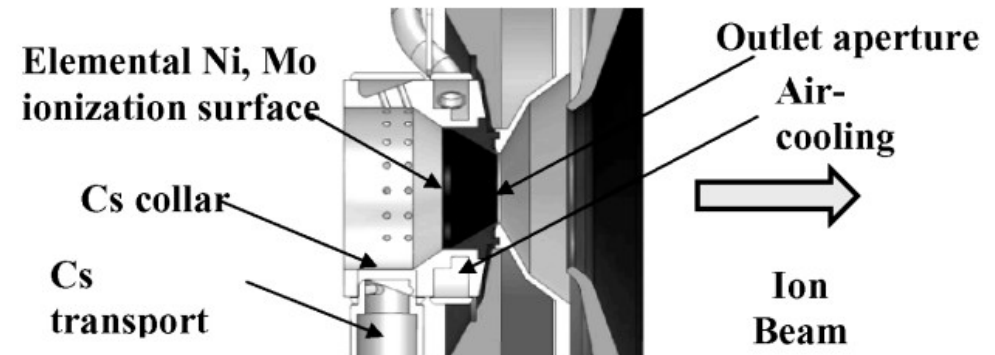
Semi-empirical mathematical relationships for electropositive adsorbate induced work function changes ☆

G.D. Alton^a

Work function

Surface	Work function [eV]	Minimum work function [eV]
Stainless steel	4.3 [56]	1.68
W	4.55 [56]	1.62
Mo	4.6 [56]	1.60
Re	4.95 [56]	1.52
Ni	5.15 [56]	1.47
Pd	5.12 [56]	1.48
Pt	5.65 [56]	1.35
W + H ₂	5.05 [59]	1.50
Ni + H ₂	5.25 [59]	1.45

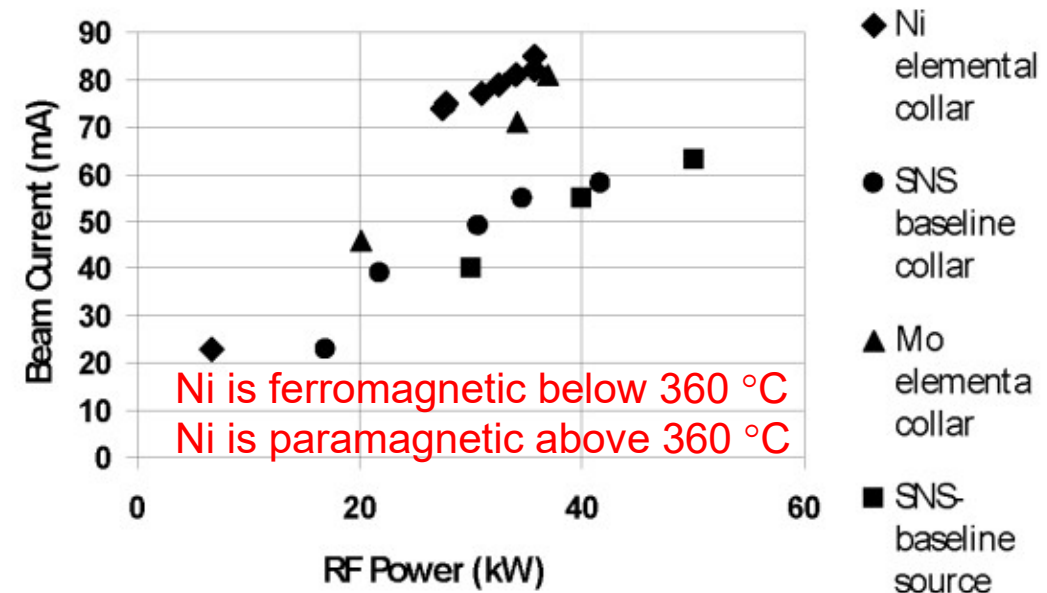
Known catalysts



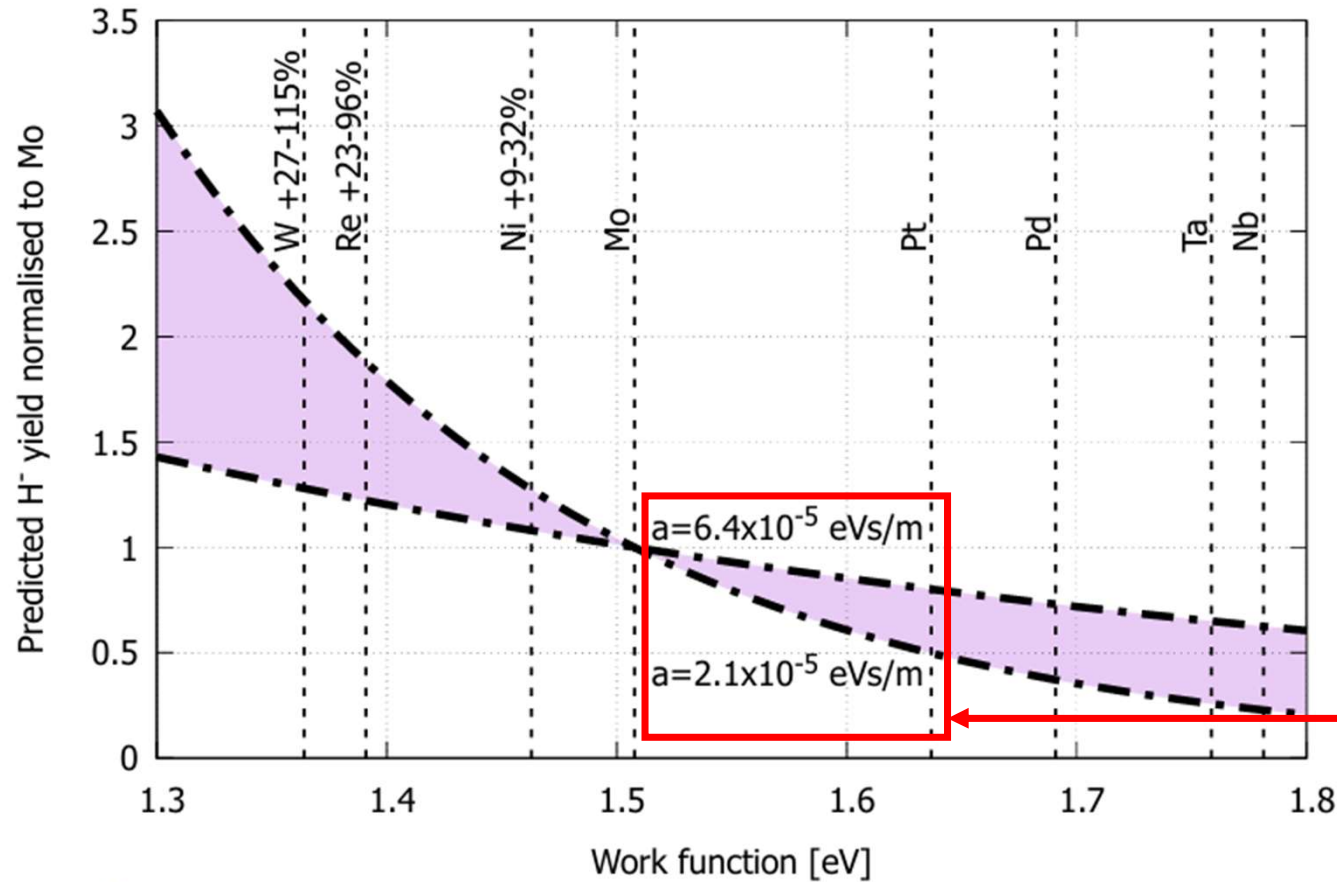
Next Generation H⁻ Ion Sources for the SNS

Cite as: AIP Conference Proceedings **1097**, 181 (2009); <https://doi.org/10.1063/1.3112511>
Published Online: 23 March 2009

R. F. Welton, M. P. Stockli, S. N. Murray, et al.



Predicted H⁻ yield relative to Mo

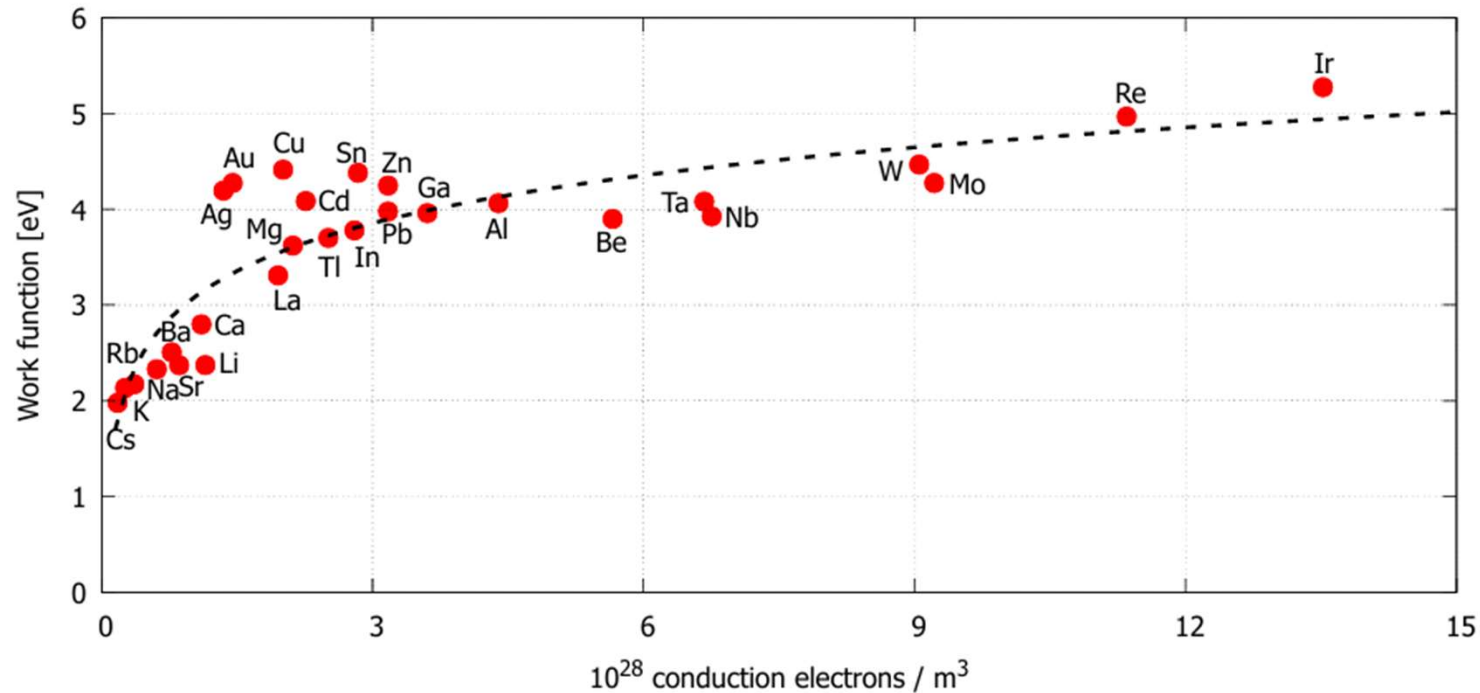


$$p_{-} \simeq \frac{2}{\pi} e^{\frac{-\pi(\phi - E_a)}{2av_z}}$$

BUT...

$$R = 2\pi \Sigma |T_i|^2 n_e$$

Conduction electron density (Fermi energy)



Density Functional Theory calculations of work function AND conduction electron density required

Surface	Conduction electron density [$\times 10^{28}$ $1/m^3$]
W - no Cs	6.4
W - Cs 0.5 monolayers	0.17
W - Cs 1 monolayer	0.23



Surface Science
Volume 55, Issue 1, April 1976, Pages 246-258



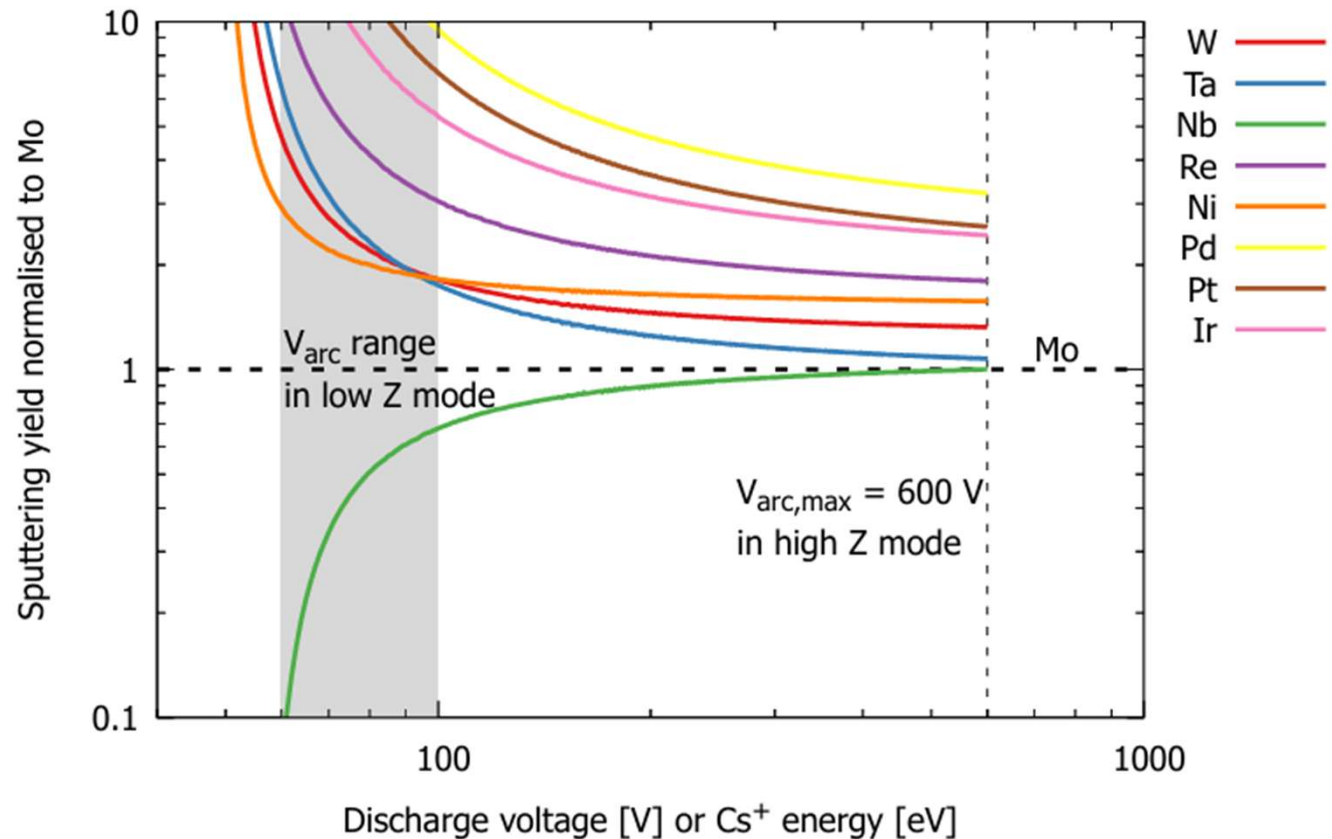
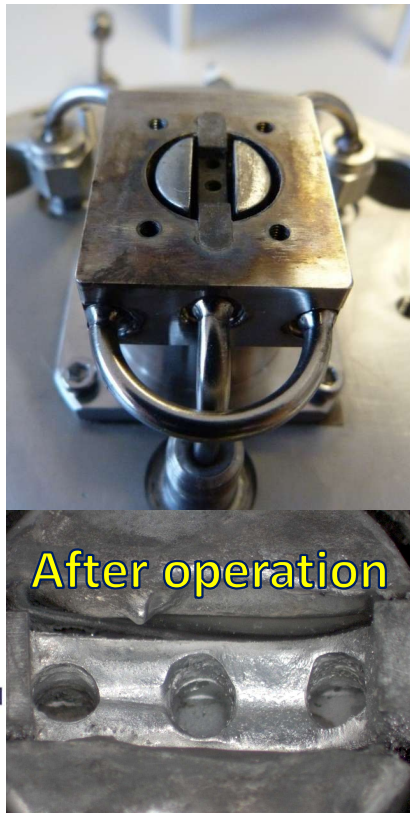
Electronic properties of alkali
submonolayers on a metal substrate

K.F. Wojciechowski *

Other material selection considerations

Material deposition: in filament-sources the surfaces becomes covered by Ta

Sputtering: unwanted deposition and lifetime of the ionisation surface



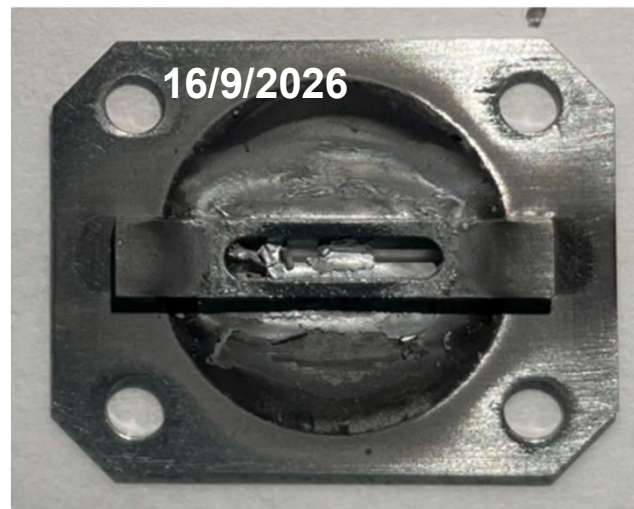
Other material selection considerations

Thermal cycling of materials facing pulsed plasmas causes **spalling** (flaking).

Molybdenum has low dynamic spall strength ($< 1\text{GPa}$) and is brittle. Internal stresses cause material to delaminate at grain boundaries.

Flake formation is a known failure mode of Penning sources (short circuit, blockages).

Potential solutions: brittle-to-ductile transition via heat treatment (800-900 °C) or Mo-Re alloy?



Recommendations

Comparison of the VUV-emission spectrum of an arc discharge and an RF discharge H^- ion source.

Measurement of the H^- density in an RF plasma near different metal surfaces at different magnetic filter fields.

Measurement of the work function in high-current ion source and comparison to DFT calculation.

DFT calculation of the conduction electron densities of caesiated metals.

Comparison of H^- beam current in high-current RF source with different converter materials (Mo, W, Re, Ni, Pt, Pd) at different surface temperatures.



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Thank you



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