The effect of addition of noble gases on negative hydrogen ion production in a dc filament discharge

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Abstract

The effect of the addition of helium, neon, argon and xenon on the production of negative hydrogen ions has been studied in a magnetically confined dc filament discharge. The addition of helium and neon produced effects similar to an equivalent increase in hydrogen pressure. However the addition of argon and low fractions of xenon produced significant increases in the negative ion density for hydrogen at pressures around 1 mTorr. The addition of argon and xenon, by increasing electron density and decreasing electron temperature, achieved conditions closer to optimum for negative ion production. The largest enhancement of negative hydrogen ion density occurred with the addition of argon; it is suggested that this is due to a resonant energy exchange between excited argon atoms and hydrogen molecules.

1. Introduction

The production of negative hydrogen ions in low pressure hydrogen discharges has been widely studied both theoretically and experimentally because of the importance of negative hydrogen ions for the production of neutral beams for heating magnetically confined fusion plasmas [1]. The main mechanism for the production of negative hydrogen ions [2] is dissociative attachment (DA) to vibrationally excited hydrogen molecules:

$$\mathrm{e} + \mathrm{H_2}(v^*) \rightarrow \mathrm{H}(1s) + \mathrm{H}^- \tag{1}$$

The vibrationally excited hydrogen molecules are themselves produced by the two-step E,V process in which a hydrogen molecule is excited by electron collision from its ground state to an excited electronic state, followed by radiative decay to a vibrationally excited level of the ground state:

$$\begin{aligned} \mathrm{e} + \mathrm{H_2}(\mathrm{X}^1\Sigma_g^+(v=0)) &\rightarrow \mathrm{H_2}(\mathrm{B}^1\Sigma_u^+, \mathrm{C}^1\Pi u) + \mathrm{e} \\ &\rightarrow \mathrm{H_2}(\mathrm{X}^1\Sigma_g^+(v^*)) + h\nu + \mathrm{e} \end{aligned} \tag{2}$$

The major loss process, with a threshold energy of 0.75 eV, is detachment by electron collisions:

$$\mathrm{H}^- + \mathrm{e} \rightarrow \mathrm{H} + 2\mathrm{e} \tag{3}$$

In this paper we report observations of the effect on the negative hydrogen ion density of adding helium, neon, argon and xenon to a dc filament discharge in hydrogen.

2. The Experiment

The plasma source is a magnetically confined dc filament discharge as shown in Figure 1. The tungsten filaments, connected in series, are heated by a dc current of up to 80 A and are biased to -70 V with respect to the the grounded 200 mm diameter stainless steel chamber. For a distance of 300mm from the filament end, the chamber is surrounded by 12 permanent magnet columns arranged with alternating poles facing the discharge in order to produce magnetic cusps within the plasma.

Electron density (n_e), electron temperature (T_e), ion density (n_i), plasma potential (V_p), floating potential (V_f), and the electron energy distribution function (EEDF) were measured using a Scientific Systems SmartProbe Langmuir probe system [3].

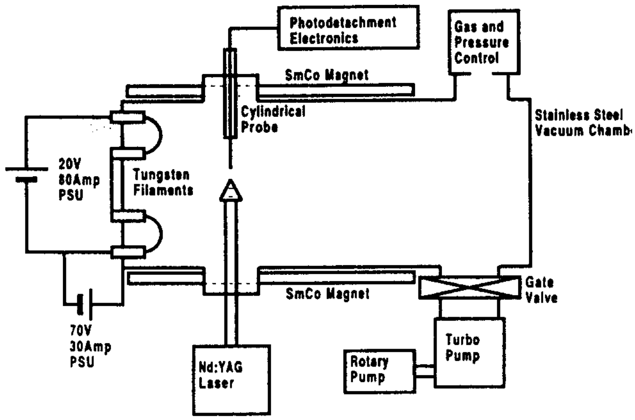

Figure 1: Schematic diagram of the filament plasma source

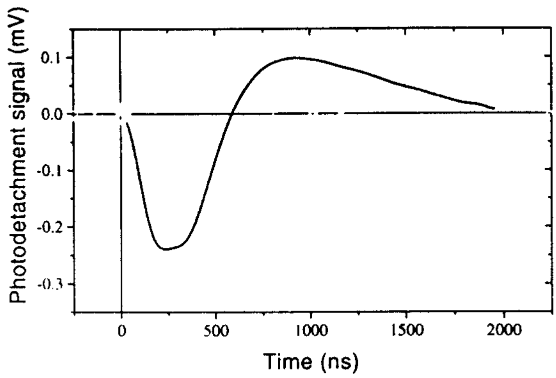

Figure 2: An example of a typical photodetachment signal

The negative hydrogen ion density was measured using a photodetachment technique due to Bacal and Hamilton [4]. A beam of photons from a Nd-YAG laser operating at its fundamental wavelength of 1064 nm removes the attached electron from the negative hydrogen ions. The photon energy, 1.17 eV, is well in excess of the electron binding energy of 0.75 eV.

By measuring the local electron density with a positively biased cylindrical probe aligned along the axis of the laser beam the electron density increase due to the photodetached electrons is detected. The change in probe electron current, ΔI^- is proportional to the electron density increase, Δn_e. Thus n_-/n_e, the ratio of negative ion to electron density, can be determined provided the unperturbed electron density is known. The negative ion density is given by

$$\frac{n_-}{n_e} = \frac{\Delta n_e}{n_e} = \frac{\Delta I}{I_e} \tag{4}$$

where I_e is the unperturbed value of the current collected by the probe. A typical photodetachment signal is shown in Figure 2.

Negative hydrogen ion densities were measured for hydrogen pressures in the range 1-5 mTorr and for a discharge current of 1 A (corresponding to a discharge power of 70 W). Measurements were also made with noble gases (helium, neon, argon, xenon) added to the hydrogen. Two series of measurements were made with the partial pressures of the added gases equal, respectively, to 20% and 40% of the hydrogen

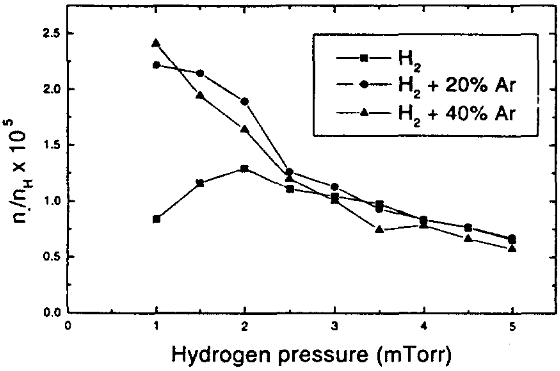

Figure 3: Relative negative hydrogen ion density n_-/n_H as a function of hydrogen pressure for pure hydrogen and added partial pressures of argon

Table 1: Effects of adding 20% noble gases

	$n_- \times 10^7$ (cm^{-3})	$n_-/n_H \times 10^6$	$n_e \times 10^{-9}$ (cm^{-3})	T_e (eV)
H_2	5.3	8.2	4.0	4.5
H_2 + 20% H_2	5.5	8.6	4.8	4.0
H_2 + 20% He	5.6	8.7	3.8	4.0
H_2 + 20% Ne	6.4	10	4.6	4.8
H_2 + 20% Ar	14	22	9.4	3.3
H_2 + 20% Xe	13	20	33	2.1

pressure.

3. Results

As an example, the results obtained with added argon are shown in Figure 3. The ratio of negative hydrogen ion density to the equivalent hydrogen atom density at the filling pressure (n_-/n_H) is shown as a function of hydrogen pressure. For 20% added noble gas, probe measurements of electron density and electron temperature are shown in Figures 4 and 5 respectively.

4. Discussion

The effect of adding noble gases generally increases with the atomic mass of the gas added. The effect on the electron temperature, electron density, and negative ion density of adding helium and neon is not significant. Argon however produces a large increase in electron density, a decrease in electron temperature, and a significant increases in the relative efficiency of negative ion production (n_-/n_H) at the lower end of the pressure range studied. The addition of xenon caused a very large increase in electron density and significant lowering of the electron temperature; 20% Xe produced an increase in the relative efficiency of negative ion production (n_-/n_H) at the lower pressures studied; there is, however, little effect for 40% Xe.

The effects of adding noble gases to a 1 mTorr hydrogen discharge to increase the pressure by 20% and 40% are summarised in Table 1 and Table 2 respectively. For comparison, results corresponding to a 20% and 40% increase in the pressure of the pure hydrogen discharge are also given. It is clear that the effects of adding helium or neon are similar to those achieved by adding the same amount of hydrogen, while the more dramatic effects of adding argon and xenon are clearly apparent.

As the threshold for the E, V process for the production of excited vibrational states of H_2 is 11.37 eV [5], the electrons responsible for vibrationally excited H_2 are not significantly affected by the addition of argon or xenon since the high energy part of the measured EEDF shows little change upon the addition

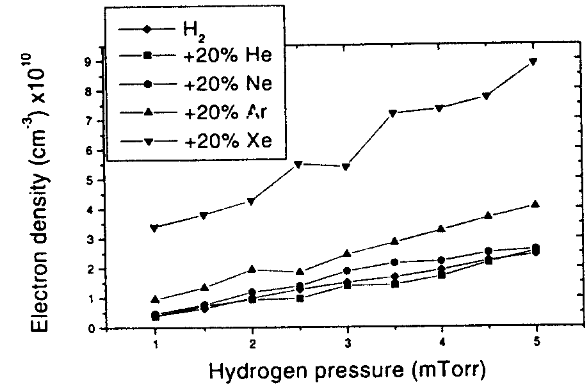

Figure 4: Electron density as a function of hydrogen pressure for pure hydrogen and added helium, neon, argon and wenon corresponding to pressure increases of 20%

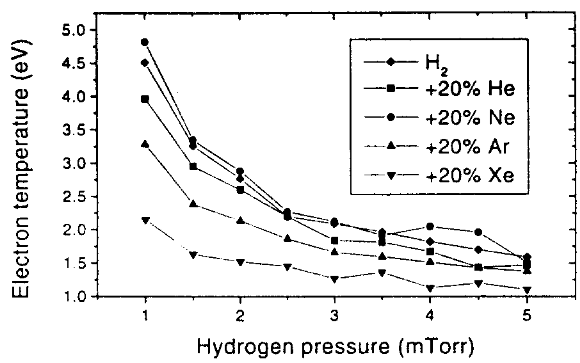

Figure 5: Electron temperature as a function of hydrogen pressure for pure hydrogen and added helium, neon, argon and wenon corresponding to pressure increases of 20%

Table 2: Effects of adding 40% noble gases

	$n_- \times 10^7$ (cm^{-3})	$n_-/n_H \times 10^6$	$n_e \times 10^{-9}$ (cm^{-3})	T_e (eV)
H_2	5.3	8.2	4.0	4.5
H_2 + 40% H_2	9.0	14	6.0	3.5
H_2 + 40% He	7.2	11	4.2	3.9
H_2 + 40% Ne	9.1	14	5.3	4.5
H_2 + 40% Ar	15	24	14	2.6
H_2 + 40% Xe	8.8	14	57	1.6

of the noble gas. However, the subsequent DA process which leads to the production of H^- ions has an increasing cross-section and decreasing threshold with increasing vibrational level v. For example the cross-section near threshold increases by five orders of magnitude between $v = 0$ and $v = 7$ while the threshold energy falls from 3.75 eV to less than 1 eV [6]. The DA process can therefore be expected to be sensitive to variations in electron temperature.

Calculations of the dissociative attachment reaction rate show that it peaks slightly above 1 eV and that the electron detachment loss rate increases with electron temperature in the range of interest. This suggests that the enhanced H^- density resulting from the addition of argon or low fractions of xenon at the low pressure end of the measurements is due to an increase in electron density (since both argon and xenon are more easily ionised that helium, neon or hydrogen itself) accompanied by a lowering of electron temperature to bring it much close to the optimum for dissociative attachment and at the same time to reduce the electron detachment rate. As pressure is increased the relative effects on electron density and temperature of adding argon and 20% xenon decrease, giving results little different from the addition of helium, neon or hydrogen itself.

We believe that there is a second mechanism occuring in the argon seeded plasma which is due to resonances between excited states in argon and hydrogen molecules. The energies of the lowest (1s) excited states for helium lie around 20 eV above the ground state; these energies are around 17 eV for neon, 12 eV for argon and 9 eV for xenon [7]. The energy required to excite a hydrogen molecule in the ground vibrational level ($v = 0$) electronic state is in the range 11.18-12.02 eV for v in the range 0-5.

For argon (but not for helium, neon or xenon) there are near coincidences with excited vibrational states of the first excited electronic state.

There are two possible ways that these energy coincidences between argon atoms and hydrogen molecules can lead to an increase in the density of vibrationally excited H_2 in the discharge. One possibility is that collisions between metastable argon atoms($1s_3, 1s_5$) and ground state H_2 can efficiently generate excited molecules. This collision process has been studied [8] and although it does produce excited molecules, they are in the $a^3\Sigma_g^+$ state which decays into the dissociative $b^3\Sigma_u^+$ state so the ultimate products of this reaction are hydrogen atoms. Collisions with metastables also form the ArH excimer molecule [9].

The other possibility is a sequential energy transfer from the argon states via a photon:

$$e + Ar \rightarrow Ar(1s_4, 1s_2) \rightarrow Ar + h\nu \tag{5}$$

followed by

$$h\nu + H_2 \rightarrow H_2(B^1\Sigma_u^+(v^*), C^1\Pi_u(v^*)) \rightarrow H_2(X^1\Sigma_g^+(v^*)) + h\nu' \tag{6}$$

where $\nu \neq \nu'$. The argon states involved are dipole allowed and have large excitation cross sections in comparison with the metastable states. Furthermore, the radiation is trapped in the discharge so excitation of hydrogen molecules may be the main loss process. Experimental evidence that this can be an important energy transfer process was obtained by Lyman [10], and studied more recently by Takezawa *etal* [11].

Thus in the argon and hydrogen mixtures the increase in the population of the vibrationally excited hydrogen molecules, $H_2(v^*)$, together with the lower electron temperature and increased electron density leads to a significant increase of the negative ion density.

5. Conclusions

It has been found that the addition of argon and xenon to a magnetically confined dc filament discharge in hydrogen produces an increase in the efficiency of negative hydrogen ion production for hydrogen pressures of ~ 1 mTorr. In contrast, the addition of helium or neon produces effects which are negligibly different from those due to an equivalent significant lowering of electron temperature to bring it closer to the optimum value for negative ion production of ~ 1 eV. A resonant energy transfer between excited argon atoms and hydrogen molecules leads to an increase in vibrationally excited hydrogen molecules and consequently further enhancement of the negative ion density in the argon and hydrogen mixtures.

Acknowledgements

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Brian William James was born in Sydney, Australia in 1946. He received a BSc with first class honours in physics from the University of Sydney in 1967 and a PhD in experimental plasma physics in 1971. His research interests have included ionising shock waves, plasma centrifuges, submillimetre laser development, coherent scattering from high temperature plasmas, and low temperature plasmas and their applications. His present interests are centred on laser and spectroscopic diagnostics of plasmas. His research career includes visiting postions at the Culham Laboratory, UCLA, Kyushu University, and Dublin City University. He is presently an associate professor in the School of Physics at the University of Sydney and an associate dean of the Faculty of Science.