Comparison of measured and modelled negative hydrogen ion densities at the ECR-discharge HOMER

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Comparison of Measured and Modelled Negative Hydrogen Ion Densities at the ECR-Discharge HOMER

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Abstract.
As the negative hydrogen ion density n_{H^-} is a key parameter for the investigation of negative ion sources, its diagnostic quantification is essential in source development and operation as well as for fundamental research. By utilizing the photodetachment process of negative ions, generally two different diagnostic methods can be applied: via laser photodetachment, the density of negative ions is measured locally, but only relatively to the electron density. To obtain absolute densities, the electron density has to be measured additionally, which induces further uncertainties. Via cavity ring-down spectroscopy (CRDS), the absolute density of H^- is measured directly, however LOS-averaged over the plasma length. At the ECR-discharge HOMER, where H^- is produced in the plasma volume, laser photodetachment is applied as the standard method to measure n_{H^-}. The additional application of CRDS provides the possibility to directly obtain absolute values of n_{H^-}, thereby successfully benchmarking the laser photodetachment system as both diagnostics are in good agreement. In the investigated pressure range from 0.3 to 3 Pa, the measured negative hydrogen ion density shows a maximum at 1 to 1.5 Pa and an approximately linear response to increasing input microwave powers from 200 up to 500 W. Additionally, the volume production of negative ions is 0-dimensionally modelled by balancing H^- production and destruction processes. The modelled densities are adapted to the absolute measurements of n_{H^-} via CRDS, allowing to identify collisions of H^- with hydrogen atoms (associative and non-associative detachment) to be the dominant loss process of H^- in the plasma volume at HOMER. Furthermore, the characteristic peak of n_{H^-} observed at 1 to 1.5 Pa is identified to be caused by a comparable behaviour of the electron density with varying pressure, as n_e determines the volume production rate via dissociative electron attachment to vibrationally excited hydrogen molecules.

INTRODUCTION

For the investigation of negative ion sources, the negative hydrogen ion density n_{H^-} is a key parameter. Therefore its reliable diagnostic quantification is an essential task concerning source development and operation as well as regarding fundamental research. Diagnostic accessability to this crucial parameter can particularly be provided by utilizing the photodetachment process of H^-. Thereby the additional electron is detached from the negative ion by a photon, on condition that its energy $h\nu$ is above the binding energy of the electron of 0.75 eV. Experimentally, this process can be induced by irradiating laser light into a hydrogen plasma, which forms the basis of two different diagnostic approaches.

Via *laser photodetachment* the photodetached electrons are detected as an increase of the electron current measured by a Langmuir probe system. This diagnostic method has been investigated particularly by Bacal et al. [1, 2]. On the one hand, it allows for local measurements of the negative ion density and thus for space-resolved investigations, e.g. the determination of spatial n_{H^-} profiles. On the other hand, the negative ion density is detected relatively to the electron density of the plasma. In order to obtain absolute values of n_{H^-}, the electron density has to be determined separately, possibly leading to additional uncertainties. Furthermore, laser photodetachment is an invasive diagnostic method which may have an influence by inserting and biasing the probe. In general the experimental application of laser phototdetachment is demanding, and the reliability of the obtained negative ion density has to be investigated for each diagnostic setup individually.

Alternatively, negative hydrogen ions can also be quantified by detecting the absorption of the laser light due to the photodetachment process. This approach is pursued via (pulsed) *cavity ring-down spectroscopy (CRDS)*, where the decay time of a laser pulse in an optical cavity is measured. This high-sensitive method of absorption spectroscopy was introduced by O'Keefe and Deacon [3] and since then frequently applied to obtain reliable absolute, but line-of-sight averaged negative hydrogen ion densities [4, 5, 6, 7]. In contrast to laser photodetachment, CRDS is a non-invasive diagnostic method, but not capable of specifically space-resolved investigations of the negative ion density. However, its ability to directly obtain absolute values of n_{H^-} provides the beneficial possibility to benchmark the ion densities

obtained by laser photodetachment, when both diagnostic methods are simultaneously applied and compared.

Such investigations of the negative ion density are conducted at the small-scale ECR test bed HOMER (**HOM**ogeneous **E**lectron cyclotron **R**esonance plasma). Therefore the experiment is operated in hydrogen at a pressure range from 0.3 to a few Pa. To reduce the destruction of negative ions by electrons in the plasma volume, HOMER is operated as a tandem source: via the application of a meshed grid, the plasma is divided into a heated, and a diffusive, non-heated part. In the latter, the losses of negative ions due to electron detachment are damped as the electron density and temperature are reduced [8]. In this downstream section of HOMER, laser photodetachment is applied as the standard diagnostic method to measure the density of negative ions. In order to benchmark this system, CRDS is installed additionally and measurements of n_{H^-} are conducted in the pressure range from 0.3 to 3 Pa and at varying microwave powers between 200 and 500 W. Particular experimental measures to protect the cavity mirrors from plasma-caused degradation, which have to be adopted for reliable and sensitive density measurements via CRDS, are presented as well.

Furthermore, a 0-dimensional model of the volume production of H^- is applied in order to identify the dominant destruction processes of negative ions in the plasma volume at HOMER. By balancing the production rate via dissociative electron attachment to vibrationally excited hydrogen molecules with the relevant loss processes caused by collisions of H^- with plasma particles, the density of negative hydrogen ions is calculated based on various measured plasma parameters. For the comparison and verification of the modelled densities the absolute density measured by CRDS is a suitable parameter. By adapting the modelled densities to the measurements of CRDS, further insight of the volume production of H^- via dissociative electron attachment at HOMER is possible.

EXPERIMENTAL SETUP AND DIAGNOSTIC METHODS

A schematic of the ECR-setup HOMER is shown in figure 1. It consists of a cylindrical plasma vessel of 15 cm in diameter and 31 cm in height. At a frequency of 2.45 GHz a microwave is generated by a magnetron with a maximum output power of 1 kW, entering the vessel through a dielectric window. The required magnetic field of 87.5 mT is created by two water-cooled coils. The floating meshed grid (stainless steel) prevents the expansion of the microwave into the lower part of the vessel and thereby separates the plasma volume into a heated section above (height 12 cm) and a non-heated, diffusive section (height 19 cm) below it. Especially the destruction of negative ions due to electron detachment in the downstream section below the grid is thereby reduced. Dedicated investigations regarding the influence of the meshed grid on the plasma parameters at HOMER are given in [8]. In the downstream section four diagnostic ports are available for the simultaneous installation of both CRDS and laser photodetachment. Additionally, measurements via Langmuir probe as well as optical emission spectroscopy (OES) can be performed to derive further plasma parameters. At the bottom plate, a height adjustable sample holder is installed, which allows for the investigation of the influence of different sample surfaces on the plasma parameters, especially on the negative ion density [8]. For reference and in order to ensure the comparability of the plasma geometry with such investigations, a stainless steel sample is installed for the presented measurements.

For both diagnostic methods presented in the following, the required laser light to induce the photodetachment process is provided by pulsed Nd:YAG-lasers emitting at a wavelength of 1064 nm. The exact layout of both lines-of-sight (LOS) is displayed on the lower part of figure 1. Seen from the top the LOS are arranged perpendicular to each other and in a vertical distance of 1 cm. For the comparability of both diagnostics, this difference in height is not critical. Due to the applied homogeneous magnetic field, the plasma parameters at HOMER have the distinction of being spatially homogeneous, especially in the vessel centre.

Laser photodetachment

Laser pulses of 8 ns length and a beam diameter of about 6 mm are injected into the plasma, passing through a quartz window. At the typical pulse energies of 25 mJ all negative hydrogen ions are detached inside the LOS of the laser, which is adjusted concentrically on the tip of a Langmuir probe. The latter consists of a tungsten wire (length 1 cm, diameter 100 µm), which is brought into the plasma via a stainless steel rod inside an insulating glass tube. The probe is positively biased ($U = +35$ V) and thereby operated in the electron saturation regime, where the measured current is solely carried by the plasma electrons. When a laser pulse is injected, the additional electrons of the negative hydrogen ions are detached and collected by the positively biased probe, leading to a sharp increase of the measured electron
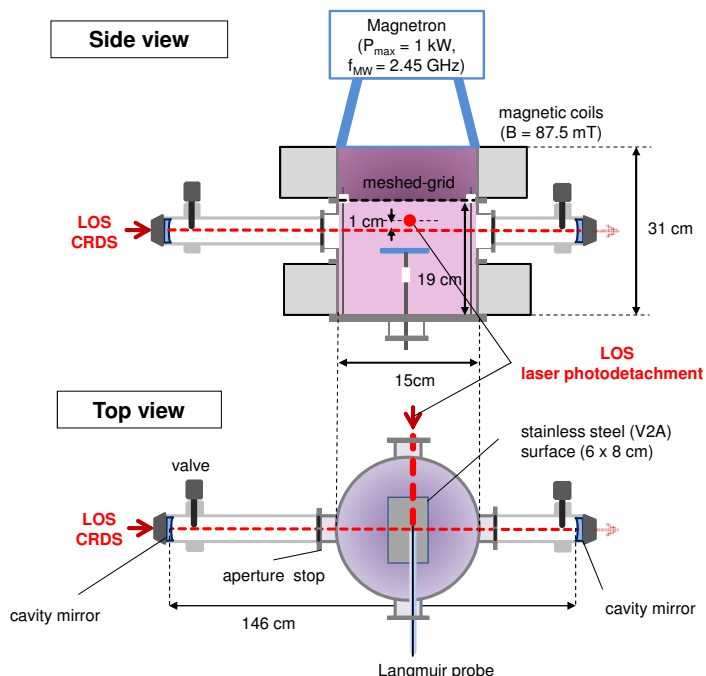
FIGURE 1. Schematic of the ECR-setup HOMER. Top and side view illustrate the arrangement of both laser photodetachment and CRDS.

current for about 1 µs. By measuring the amplitude of this increase, the local density of negative hydrogen ions can be obtained, relatively to the electron density of the plasma [1]. In order to significantly increase the signal-to-noise ratio, typically 15 single shots are averaged. In order to acquire absolute values of the negative ion density n_{H^-}, the electron density n_e has to be determined by Langmuir probe measurements additionally: assuming quasineutrality, n_e is obtained by measuring the positive ion density in the ion saturation regime.

Cavity ring-down spectroscopy

Laser pulses of 7 ns length and 3 mm beam diameter are injected into an optical cavity consisting of two axisymmetrically arranged, planar-confocal and highly reflective mirrors (distance: 1.46 m, curvature: 1 m, $R = 99.994\%$). The pulse is reflected back and forth between the mirrors, with a small fraction of light transmitting through them with each pass. The temporal behaviour of the transmitted light, which is observed by a photo diode detector, resembles an exponential decay characterized by a typical *intrinsic* decay time. With the present setup intrinsic decay times of 75 µs are achieved. If a hydrogen plasma is generated inside the cavity, additional absorption losses due to the photodetachment of H$^-$ occur. Consequently, the decay time is reduced. The high sensitivity of this method is a direct result of the pulse passing the plasma multiple times, virtually increasing the absorption length. By separately measuring, on the one hand, the intrinsic decay time in vacuum/gas phase and, on the other hand, the decay time during the plasma phase, the density of negative hydrogen ions can be quantified [3]. The obtained densities are absolute, but averaged over the absorption length which is in this case the plasma diameter (15 cm). Consequently, CRDS is not capable of specifically space-resolved measurements, in contrast to laser photodetachment. Apart from the intrinsic uncertainties of the measured decay times and the required value of the photodetachment cross section [9], no additional errors can influence the measurement, as CRDS requires no further plasma parameters to obtain n_{H^-}.

However, for a successful application of CRDS at HOMER, several issues have to be regarded. First of all, the typical negative ion densities at HOMER are of the order of $10^{15}\,\mathrm{m^{-3}}$. Therefore, the system has to be sensitive

to variations of the order of $10^{14}\,\text{m}^{-3}$ to conduct systematic investigations. This imposes requirements for both the mirror reflectivity, which defines how many times the plasma is passed by the laser pulse, and the accuracy of the measured decay times, which defines the smallest detectable difference between the intrinsic and the plasma decay time. At the given mirror reflectivity of $R = 99.994\,\%$, the required sensitivity for n_{H^-} can be achieved by averaging 50 acquisitions of both decay times, thereby reducing the statistical errors of the decay time measurement. Furthermore, mechanical vibrations at the test bed (e.g. caused by pumping and cooling systems) have to be damped sufficiently, as they have shown to cause a periodic misalignment of the cavity leading to additional statistical errors.

Regarding the cavity mirrors themselves, a characteristic degrading effect due to the hydrogen plasma is observed if the mirrors are integrated into the setup without any protective measures: on a time scale of few seconds up to several minutes, the reflectivity continuously decreases during plasma phases. This loss recovers only slowly and even incompletely in vacuum or gas phase afterwards. Comparable behaviour has already been reported, mostly ascribing the degradation to interactions of plasma particles with the mirror surfaces [10, 11]. However, specific investigations at HOMER revealed another mechanism to be a possible cause: plasma-emitted (V)UV radiation altering the reflectivity properties of the cavity mirrors. The spectra of low temperature low pressure hydrogen plasmas typically show significant emissions below 300 nm, due to several molecular and atomic transitions. The exposure of the cavity mirrors to radiation of this wavelength can lead to electronic defects within their dielectric coatings, such as the build-up of so-called color centres [12, 13]. These defects can significantly absorb light in the visible and IR range, thereby reducing the reflectivity.

This effect causes two very specific practical problems for the diagnostic use of CRDS: Firstly, the permanent reflectivity loss leads to a reduced sensitivity. Secondly, the reflectivity loss significantly reduces the decay time in plasma phases, thereby superimposing the drop of the decay time due to the photodetachment of H^- and preventing accurate and undisturbed measurements of n_{H^-}. Therefore, experimental measures are mandatory in order to protect the mirrors from the degrading influence of the plasma. Valves are applied in order to completely isolate the mirrors when CRDS is not actively used. During measurements, the solid angle from which UV radiation and/or particles from the plasma volume can reach the mirror surfaces is strongly reduced by applying concentric aperture stops with an inner diameter of 8 mm. Thereby the mirrors are mostly shielded, without impairing the LOS of the laser. The application of these protective measures, which are also illustrated in figure 1, successfully dampens the degradation on a time scale of a few minutes, which is sufficient for the required 50 single shots.

MEASUREMENT OF n_{H^-} AND BENCHMARK OF LASER PHOTODETACHMENT

Figure 2 shows measured negative ion densities at the ECR-discharge HOMER. Illustrated in figure 2 (a) is the behaviour of n_{H^-} in the pressure range from 0.3 to 3 Pa at varying microwave power between 200 W and 500 W as detected via CRDS. Independent from the applied power, the ion density reaches a maximum at 1 to 1.5 Pa and decreases monotonously with lower and higher pressure. With increasing microwave power, n_{H^-} shows an approximately linear behaviour. Consequently, the maximum density of $n_{\text{H}^-} = 3.9 \times 10^{15}\,\text{m}^{-3}$ is achieved at 1 Pa and a microwave power of 500 W. Further discussion regarding the pressure dependence is provided in the next section.

Figure 2 (b) illustrates the comparison of the negative ion densities measured by CRDS and laser photodetachment at a fixed microwave power of 300 W with varying pressure in a range of 0.3 to 3 Pa. Generally, the ion densities acquired via laser photodeachment are in good agreement with the ones obtained by CRDS, regarding its absolute values as well as its pressure dependency, as both characteristic error margins significantly overlap. Slight pressure-dependent deviations between both diagnostics may be caused by the difference of local and LOS-averaged measurement. Overall, this particularly demonstrates the reliability of the absolute value of the negative ion densities obtained via laser photodetachment.
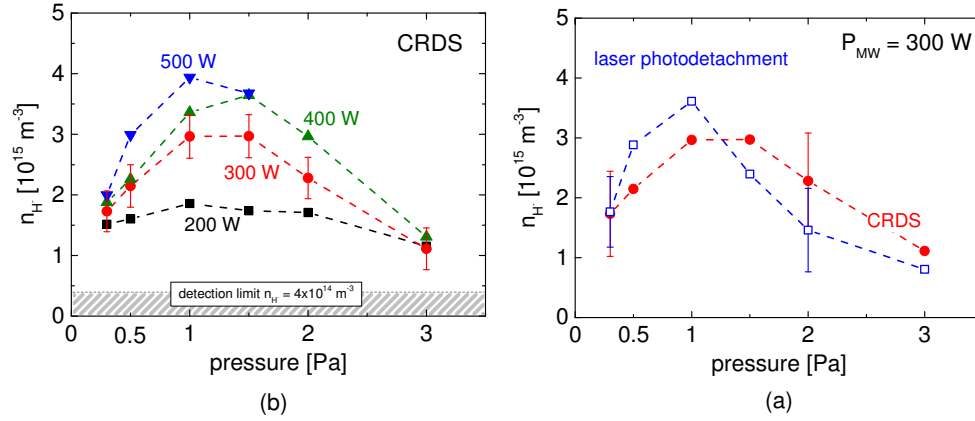
FIGURE 2. (a) Via CRDS measured negative hydrogen ion density at varying absolute pressure and different microwave powers. (b) Comparison of n_{H^-} measurements via CRDS and laser photodetachment at varying absolute pressure.

COMPARISON OF MEASUREMENT AND MODELLING

In order to identify dominant loss processes of H^- in the plasma volume at HOMER, n_{H^-} is 0-dimensionally modelled by calculating and balancing the rates of relevant production and destruction processes of negative ions. In table 1, all considered processes are listed. By applying the model, further insight into the physics behind the characteristic pressure dependency of n_{H^-} can be provided by as well.

The volume production of H^- is driven by the process of dissociative electron attachment. Thereby an electron of low energy (around 1 eV) is attached to a vibrationally excited hydrogen molecule in the electronic ground state $H_2(X,v)$. The destruction of H^- is caused by collisions with electrons, positive ions and atomic hydrogen. A detailed overview and discussion of these processes is for example provided by [14]. In order to quantify the rate of each process at HOMER, the density and temperature of every reagent of the considered processes has to be obtained diagnostically. At HOMER, these input parameters are measured by means of OES and Langmuir probe. Beyond that, rate coefficients of the considered processes are required. These are calculated based on cross section data provided by [15].

The production of negative ions via dissociative electron attachment increases strongly with higher ($v > 5$) vibrational excitation of $H_2(X,v)$ due to significantly increasing cross sections [15]. Therefore, the knowledge of the population distribution of the vibrational states in the electronic ground state is crucial for the modelling. However, via OES only the population of vibrational states with $v \leq 4$ is diagnostically accessible [16]. Assuming a Boltzmann distribution of these states, a vibrational temperature T_{vib} can be obtained. At the considered conditions at HOMER, T_{vib} is measured to be 3000 ± 500 K virtually independent of the pressure. However, this temperature is not bound to characterize the population of higher vibrational states as well. Furthermore, the effects of the applied tandem concept at HOMER have to be taken into account. In the actively heated, yet diagnostically not accessible section above the meshed grid, the electron temperature is expected to exceed the below measured values of $T_e \leq 1$ eV, as investigations

TABLE 1. Processes of negative hydrogen ions in the plasma volume considered by the applied 0-dimensional model.

Process			
Dissociative electron attachment	$H_2(X,v) + e^-$	$\rightarrow$	$H_2^- \rightarrow H^- + H.$
Electron detachment	$H^- + e^-$	$\rightarrow$	$H + e^- + e^-$
Mutual neutralisation	$H^- + H_x^+$	$\rightarrow$	$H + H_x \quad (x = 1,2,3)$
Associative detachment	$H^- + H$	$\rightarrow$	$H_2(X,v) + e^-$
non-associative detachment	$H^- + H$	$\rightarrow$	$H + H + e^-$
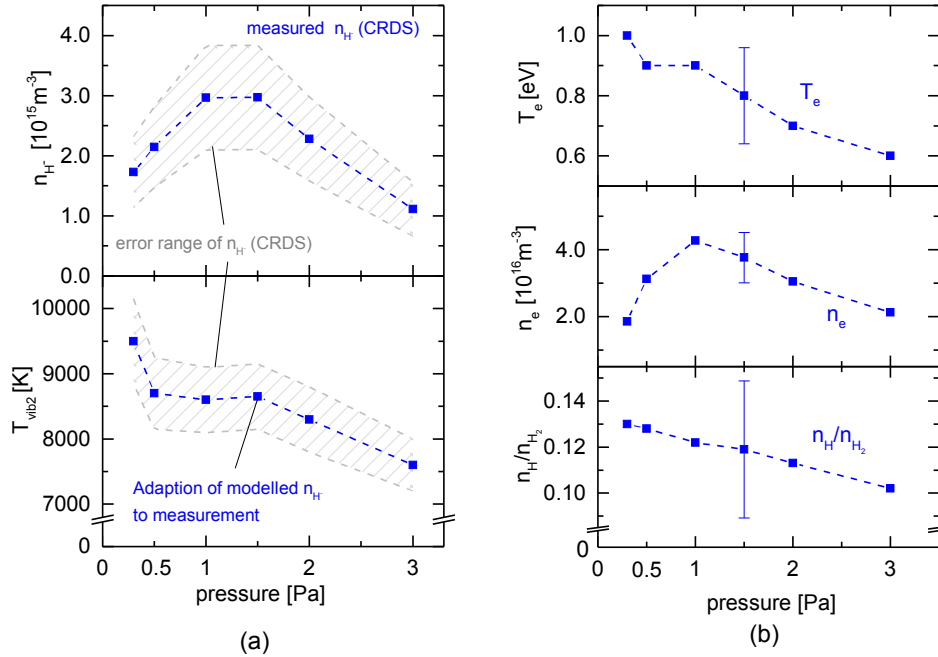
FIGURE 3. (a) Adaption of the modelled negative hydrogen ion density to the measurements of CRDS at varying absolute pressure and a microwave power of 300 W by variation of the vibrational temperature T_{vib2}. (b) Electron temperature, electron density and density ratio of atomic to molecular hydrogen at varying absolute pressure.

without the application of the meshed grid suggest [8]. This leads to the creation of highly vibrationally excited hydrogen molecules by radiative decay processes [14]. Via diffusion, these metastable particles are transported through the grid to the downstream section and effectively form H$^-$ via dissociative attachment in the region accessible to the diagnostics.

Therefore, the vibrational temperature of the upper states $v \geq 5$ can be assumed to be significantly higher than the one diagnostically obtained for $v \leq 4$. This is implemented in the model by a weighted superposition of two Boltzmann distributions, represented by two independent vibrational temperatures. The lower temperature T_{vib1} describes the population of the lower states $v \leq 4$ and is resembled by the measured value of 3000 K. The higher temperature T_{vib2} is representative for the higher states. However, as there is no diagnostic accessibility to this temperature, T_{vib2} serves as the free parameter of the modelling. By its variation, the modelled negative ion density is adapted to the measurements of CRDS. The top graph of figure 3 (a) shows the measured negative ion density at 300 W and its error margin due to the characteristic uncertainties of CRDS. In order to exactly reproduce each of these values by the model, T_{vib2} has to be varied respectively. This is illustrated in the lower graph, showing a monotonous decrease of the vibrational temperature with increasing pressure from 9500 ± 500 K to 7500 ± 500 K. Besides, the modelled densities are based on several other measured plasma parameters which show a significant pressure dependency. Those are illustrated in figure 3 (b) at varying absolute pressure between 0.3 and 3 Pa: the electron temperature, which is measured via Langmuir probe, shows a decrease from 1 to 0.6 eV with increasing pressure. The electron density, also obtained via Langmuir probe measurements, is of the order of several 10^{16} m^{-3} and shows a maximum at 1 Pa. The atomic to molecular density ratio is measured via OES and slightly decreases over pressure from 13 % to 10 %.

As the measured density n_{H^-} can be reproduced by the model, the single rates of the destruction processes of negative hydrogen ions in the plasma volume can be compared in a next step. It reveals that the collision processes of H$^-$ with H, namely associative and non-associative detachment, generally contribute to the total destruction rate with a combined share of over 90 %, due to the high density of atomic hydrogen and the low electron temperatures. Consequently, these two are the dominating destruction processes at HOMER at the considered conditions.

In a final step, the influence of the plasma parameters presented in figure 3 (b) on the production via dissociative electron attachment and the destruction via (non-)associative detachment and thus on the resulting n_{H^-} itself can be

further investigated: in the modelling, T_e determines the rate coefficients of dissociative electron attachment. However, the slightly decreasing electron temperature is shown to have no significant effect on the rate in the present case. In contrast to that, the electron density appears to have far more influence: according to the approximation that electron detachment can be neglected as a destruction proccess, n_e solely determines the volume production via dissociative attachment. Consequently, n_{H^-} is linearly proportional to n_e. Therefore the peak of n_{H^-} at 1 to 1.5 Pa is found to be a direct consequence of the analogous behaviour of n_e with increasing pressure. Finally, the slight decrease of the atomic to molecular density ratio over pressure should lead to an increase of the negative ion density due to a reduced destruction rate. This, however, can not be observed as it is superimposed and overcompensated by the reduced production of H^- due to the decreasing vibrational population represented by T_{vib2}. Conclusively, the electron density n_e and the vibrational population of the hydrogen molecule in the ground state, both determining the volume production via associative electron detachment, appear to be decisive for the observed behaviour of n_{H^-} at varying pressure.

However, these specific results are only valid for the regarded conditions at HOMER. A direct comparison to other sources such as the much larger ones of negative ion beam systems is not straightforward, as the plasma parameters influencing the considered processes may show a significantly different behaviour there and further processes may be of importance as well, for example the surface production of negative ions.

CONCLUSION

The density of negative hydrogen ions is a key parameter for the investigation and operation of ion sources and thus its diagnostic quantification is of central importance. Via laser photodetachment as well as cavity ring-down spectroscopy, the negative hydrogen ion density at the ECR test bed HOMER is measured at absolute pressures between 0.3 and 3 Pa and microwave powers between 200 and 500 W. It shows a characteristic peak at pressures of 1 to 1.5 Pa and a linear increase with the input power. Both diagnostics are in good agreement, which particularly allows for a successful benchmark of the laser photodetachment system, demonstrating its ability to reliably obtain space-resolved absolute densities as well. A 0-dimensional rate model of the volume production of H^- is applied to calculate n_{H^-} based on measured plasma parameters. Represented by a vibrational temperature, the population of the vibrational states $v \geq 5$ in the electronic ground state of the hydrogen molecule serves as the free parameter of the model. By its variation, the modelled densities can be adapted to the measurements of n_{H^-}. The processes of associative and non-associative detachment are identified to be the dominant destruction processes of H^- in the plasma volume at HOMER. As a further consequence, the characteristic, pressure dependent peak of n_{H^-} is found to be mainly caused by the electron density n_e.

ACKNOWLEDGEMENT

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