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VUV emission spectroscopy combined with H^- density measurements in the ion source Prometheus I

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Abstract. "Prometheus I" is a volume H^- negative ion source, driven by a network of dipolar electron cyclotron resonance (ECR; 2.45 GHz) modules. The vacuum-ultraviolet (VUV) emission spectrum of low-temperature hydrogen plasmas may be related to molecular and atomic processes involved directly or indirectly in the production of negative ions. In this work, VUV spectroscopy has been performed in the above source, Prometheus I, both in the ECR zones and the bulk (far from ECR zones and surfaces) plasma. The acquired VUV spectra are correlated with the negative ion densities, as measured by means of laser photodetachment, and the possible mechanisms of negative ion production are considered. The well-established H^- formation process of dissociative attachment to vibrationally excited molecules is evaluated, while an additional production path (i.e. neutral resonant ionization) is tested due to the recently attracted interest. The obtained results indicate that for the source Prometheus I, the dominant formation process is dissociative attachment.

INTRODUCTION

Volume sources of hydrogen negative ions (H^-) are considered for both fusion and accelerator applications due to many advantages, like the cesium-free operation[1]. In these sources the reaction that is widely accepted of being responsible for the production of negative ions is the dissociative attachment (DA) of low-energy (cold) electrons to molecules excited to high vibrational states[2]. While electron densities and temperatures are relatively easy to measure (e.g. with electrostatic probes), the direct experimental determination of the density of vibrational states, requires complicated diagnostics in the VUV spectral range (e.g. VUV Laser Induced Fluorescence).

These vibrational states can be formed or de-activated by various processes in the bulk plasma and on surfaces facing the plasma[3,4] (the term "bulk" stands hereafter for the plasma being far from the ECR zones and any surface; it practically refers to the region where H^- are formed). One mechanism that dominates the population of high vibrational levels, having at the same time only a small contribution to the vibrational deactivation, refers to high-energy electron impact excitation of ground state molecules to excited singlet states (e.g. $B^1\Sigma_u^+$ and $C^1\Pi_u$), followed by the spontaneous decay to high vibrational levels of the ground state[5]. Decay from the $B^1\Sigma_u^+$ singlet state gives the Lyman band, found in the 92-184 nm wavelength region, while decay from the $C^1\Pi_u$ singlet state gives the Werner band, found in the 84-158 nm wavelength region. Consequently, an indirect approach for studying the formation of vibrational states and thus the process of DA, is through the VUV emission spectrum of the discharge. In a typical VUV spectrum (Fig. 1) from the source studied here, the so-called "Prometheus I", intense Lyman and Werner band emission is observed around 160 nm and 120 nm, respectively.

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Despite the fact that DA to vibrationally excited ground state molecules is considered to be the main volume production path for negative ions, additional mechanisms should be investigated. For example, DA to the metastable $H_2(c^3\Pi_u)$[6] molecule and to molecular Rydberg states[7,8] has already been reported.

More recently, neutral resonant ionization[9] was also studied. It was proposed that the doubly excited state of $^{**}H^-(2p^2\ ^3P^e)$ is created in resonant H(2s,p) atomic collisions and there is a physical path for it to become a stable $H^-(1s^2)$ ion. A quantity that correlates well with the H(n=2) density is the Lyman-α emission intensity. Despite the intense Lyman-α radiation emitted by hydrogen plasmas (Fig. 1), the relation between negative ions and Lyman-α intensity had never been considered, until recently. Indeed, Komppula et al.[10] contributed to this topic by presenting measurements on extracted negative ions and Lyman-α intensity. By combining these data, with the elimination of the power variable, a quadratic relation between the extracted negative ion current and Lyman-α intensity was observed[9]. This relation was interpreted as an indication that negative ion formation in this source is attributed to H(n=2) - H(n=2) collisions.

Analytically, the presently accepted mechanism for collisional destruction of the H(2s) metastable hydrogen atoms is the Stark effect mixing of the metastable $2s_{1/2}$ with the neighboring $2p_{1/2}$ and $2p_{3/2}$ states of atomic hydrogen, resulting in emission of Lyman-α radiation[11]. However, collisional quenching is not the only source of Lyman-α emission. An additional source is the spontaneous emission during the 2p decay. In cases where the collisional mechanism is dominant, Lyman-α emission should be proportional to the H(2s) density (as far as opacity is negligible). Furthermore, negative ion density should be proportional to the square of the H(2s) density, according to the neutral resonance ionization mechanism. All over, a quadratic relation between negative ion density and Lyman-α emission should be established.

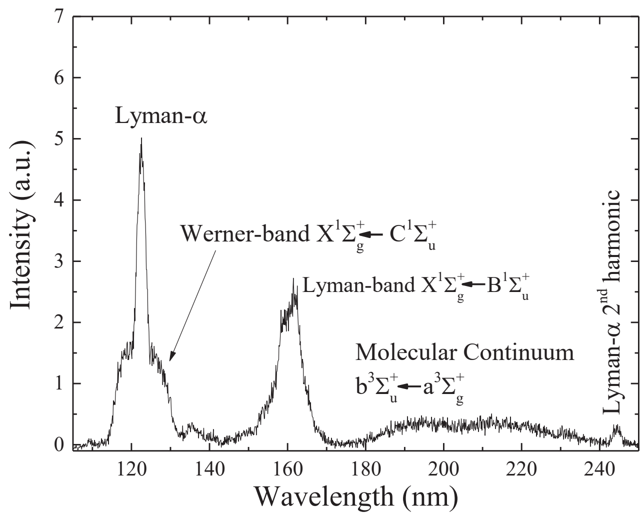

FIGURE 1. Typical VUV spectrum of the H_2 plasma in Prometheus I (12 mTorr; 180 W/module).

Apart the above three components (Lyman-α, Lyman band, and Werner band), the VUV spectrum (Fig. 1) also includes the molecular continuum emission in the 180 nm – 240 nm wavelength region. This emission corresponds to the transition from the $a^3\Sigma_g^+$ to the $b^3\Sigma_u^+$ triplet which is repulsive and leads to dissociation. Under the present working conditions, both Lyman-α intensity and molecular continuum intensity (which is a qualitative measure of the volumetric rate of the main dissociation reaction) depend linearly on the injected power[12], indicating that the increasing atomic density does not re-absorb Lyman-α emission considerably (i.e. opacity may be neglected).

In this work, the VUV spectra of the Electron Cyclotron Resonance (ECR) driven H^- source Prometheus I are recorded, and characteristic emissions are correlated with negative ion and electron densities, which are experimentally determined by means of laser photodetachment and electrostatic probes, respectively. By this way, the importance of neutral resonant ionization and DA process in H^- production may be evaluated.

EXPERIMENTAL SETUP

The negative ion source Prometheus I, schematically presented in Fig. 2, consists of a cubic stainless steel vacuum chamber having edge of 240 mm (base pressure 10^{-7} Torr; 10^{-6} Torr when the VUV monochromator is installed). It brings various viewports and feedthroughs for diagnostics. Pure H_2 is introduced with a digital mass flow controller (MKS 1179B) and the filling gas pressure is accurately monitored with an absolute pressure transducer (MKS Baratron 627D).

The plasma is generated by a network of five dipolar ECR modules[13], adapted on the top flange of the chamber. The permanent magnets of the ECR modules, also play the role of the magnetic filter of the source[14], confining hot electrons in their vicinity, while allowing both cold electrons and vibrationally excited molecules to diffuse towards the negative ion production zone. In other words, Prometheus I is designed for DA enhancement.

In this work, two cases are studied, corresponding to two different positions of the modules. In the first case, their mid-plane (i.e. the mid-plane of the permanent magnets) is positioned 203 mm above the source bottom, while in the second case, this distance is 135 mm. Each module is driven by an independent solid state power supply (2.45 GHz) able to provide up to 180 W of power. Impedance matching is accomplished with a tuner embedded on the main body of the module, allowing minimization of the reflected power (<5 W here).

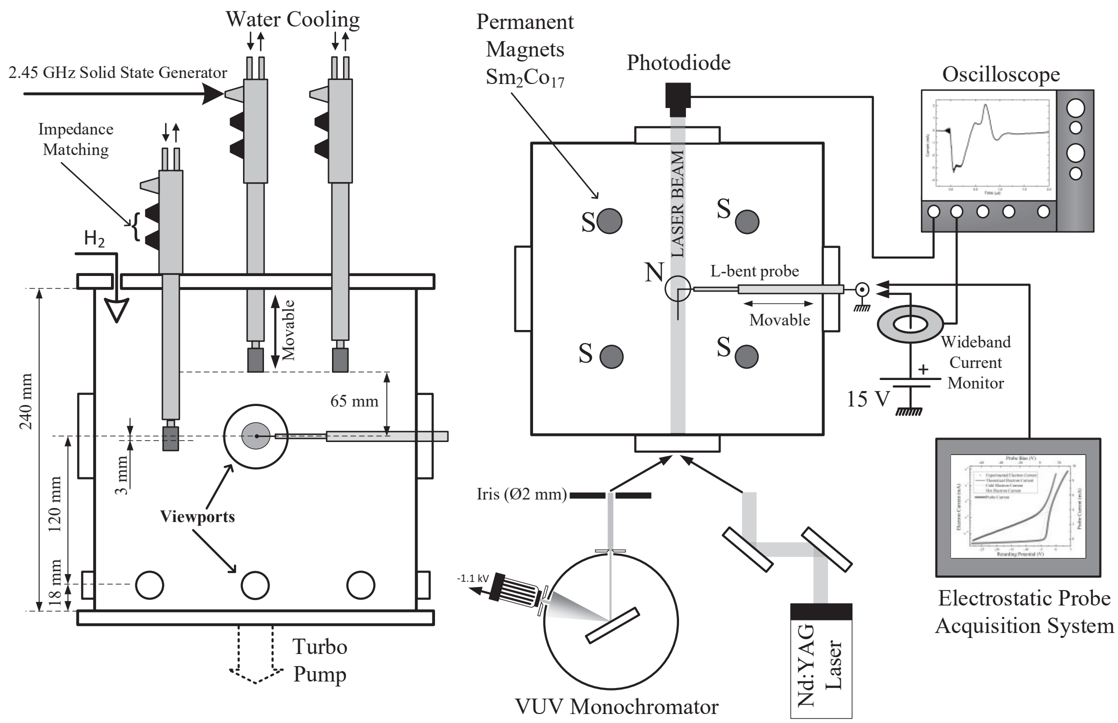

FIGURE 2. Conceptual diagram (side-view and top-view) of the source Prometheus I and the installed diagnostics: electrostatic probe, laser photodetachment, and VUV emission spectroscopy.

A cylindrical electrostatic probe is immersed laterally into the plasma, as presented in Fig. 2. The probe is made of tungsten wire, 0.25 mm in diameter. It is L-shaped for the needs of the photodetachment technique and its total length exposed to the plasma is 15 mm. The rest of the wire is housed in dielectrics, following a telescopic configuration[15]. Data are acquired and treated by a custom made system[16]. The magnetic field in the probe position is calculated to be 51 G, leading to a Larmor radius greater than the probe collecting radius. This allows the interpretation of the probe data with the classical non magnetized theory[15,17,18]. The electron energy distribution function (EEDF) in Prometheus I, exhibits a bi-Maxwellian form due to a dense low-temperature bulk electron population and a lower-density hot electron population. The experimental EEDF is derived from the second derivative of the total probe current (Druyvesteyn method)[15]. For noise reduction, a sliding Savitzky-Golay filter[19] is designed. In order to correctly apply the Savitzky-Golay filter, evenly spaced data are acquired through an interpolation procedure on the experimentally derived curve. The window of the filter starts from a constant low value (~0.3 eV) in the low energy area, and is gradually widened up to 0.3 eV per eV of electron energy[20]. Naturally, this filtering procedure reduces the resolution of the acquired EEDF for higher energies.

For the laser photodetachment technique[21], a Nd:YAG 1064 nm laser (Quantel Brilliant Eazy; 330 mJ pulse^{-1}; 5 ns pulse) is employed. The photo-detached electrons are collected by the positively biased probe and the resulting impulse is recorded by means of wideband current transformer[22] (Pearson Electronics; model 6585). The density of negative ions can then be calculated from the characteristic peak of the current impulse[21,23]. For the valid application of the technique the system parameters are chosen appropriately; a laser energy density of 70 mJ cm^{-2} ensures electron detachment from all H^- ions in the irradiated volume and a +15 V probe DC bias ensures that all the detached electrons are collected (typical plasma potential +7 V). Any error bars in the graphs below, arise from two series of measurements on different dates.

The VUV spectrometer consists of a monochromator with a focal length of 0.2 m (McPherson Model 234/302), equipped with a holographic grating and a photomultiplier tube (ET Enterprises 9406B). The spectrometer slit is combined with an extra iris to collimate the emitted light and thus increase the spectral resolution. Typical spectra are given in Fig. 3. Fig. 3a refers to the VUV wide scan spectrum from both bulk plasma (ECR modules placed at the upper position) and ECR zones (ECR modules placed at the lower position) (see as well Fig. 2 for position identification). Intense emission of the Lyman band is observed in the 142-172 nm range, while the integral in these limits is assumed here as a measure of the total Lyman band intensity. The Werner band overlaps the Lyman-α line and it is here extracted as a Gaussian approximation. This deconvolution is depicted in Fig. 3b, for indicative power levels.

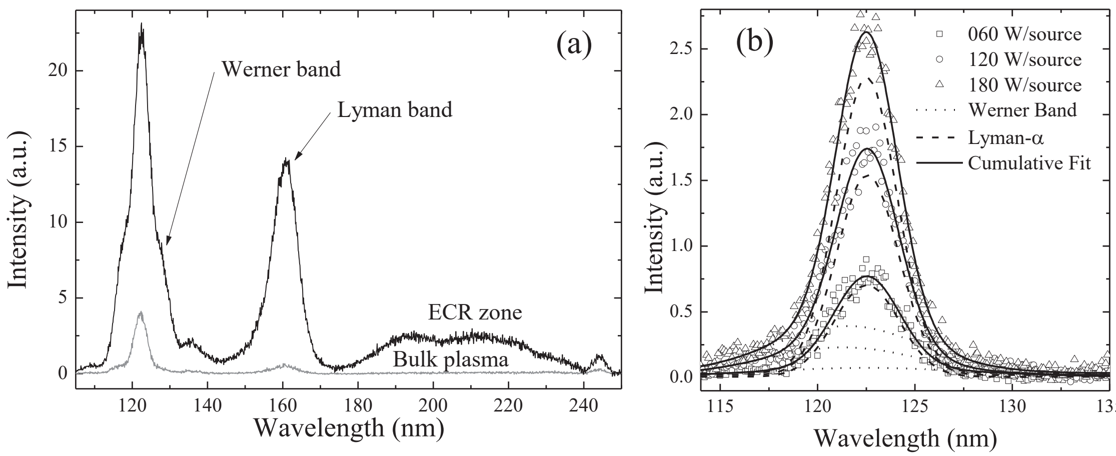

FIGURE 3. (a) Comparison of VUV emission spectra from the bulk plasma and ECR zones.
(b) Deconvolution of Lyman-α emission from Werner band.

RESULTS AND DISCUSSION

In this section, the results are presented and discussed on the base of 4 mTorr, as an indicative working pressure, without losing their generality; see Aleiferis et al.[24] for other pressures.

Lyman band intensity (integral over 142-172 nm), both from the bulk plasma and the ECR zones, is monotonically increasing in the entire power range studied here, as Figs. 4a and 4b show, respectively. It seems that the injected power can be very efficiently deposited on singlet excitation processes, leading to vibrationally excited molecules. This fact plays a significant role in DA, since the emission from the Lyman band probed here corresponds to the light emitted during the formation of highly vibrationally excited molecules[25].

Especially, the Lyman band intensity inside the ECR zones is almost two orders of magnitude higher than in the bulk plasma (relative values), demonstrating the high yield of vibrationally excited molecules in these zones. These molecules, having long lifetimes and no charge, are free to escape from the ECR magnetic filters and diffuse in the bulk plasma where the production of negative ions takes place.

Cold electrons created by ionization processes inside the ECR zones can diffuse towards the bulk plasma as well. The EEDFs (see below) in this bulk plasma region consist mainly of a cold electron population with density around

10^9 cm^{-3} and temperature ~1 eV. On the contrary, hot electrons (temperature ~15 eV) constitute less than 5% of the total electron density in this region. These conditions are quite favorable for the production of negative ions by DA.

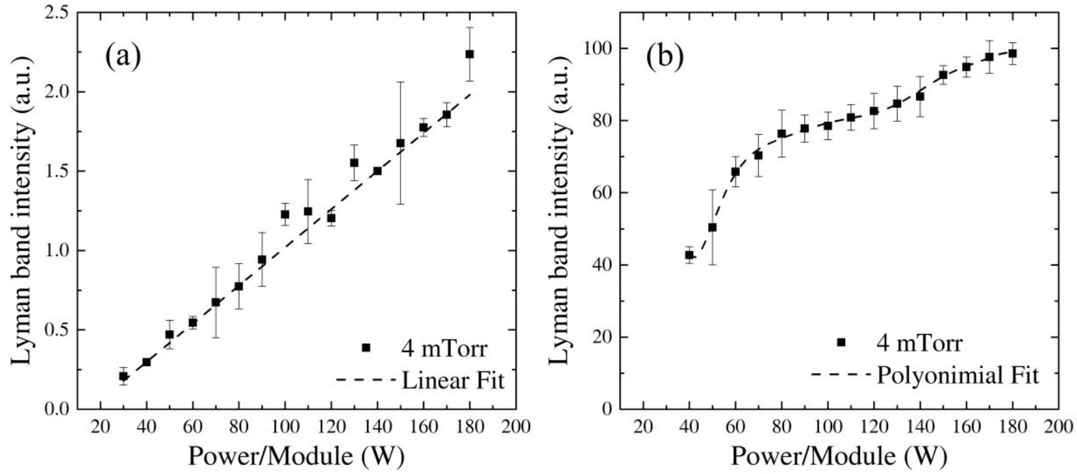

FIGURE 4. Power dependence of Lyman band emission; (a) Bulk plasma (b) ECR zones.

The bulk electron density versus power (Fig. 5a) does not monotonously increase but peaks around 120 W/module. On the other hand, Lyman band emission, as induced by $B^1\Sigma_u^+$ excitation, increases almost linearly with the power (Fig. 4a). This implies that, over the high power range studied here, the energy deposition by the hot electrons on singlet excitations instead of ionization is favored; details are provided in our previous work[12]. This fact might lead to reduced ionization rate and consequently to the decreasing electron density observed in Fig. 5a. Similar issue has been observed in other ECR sources[26], i.e. the high energy of hot electrons was more efficiently deposited on singlet excitation instead of ionization process, whereas the opposite effect was observed in a filament discharge H^- source.[10]

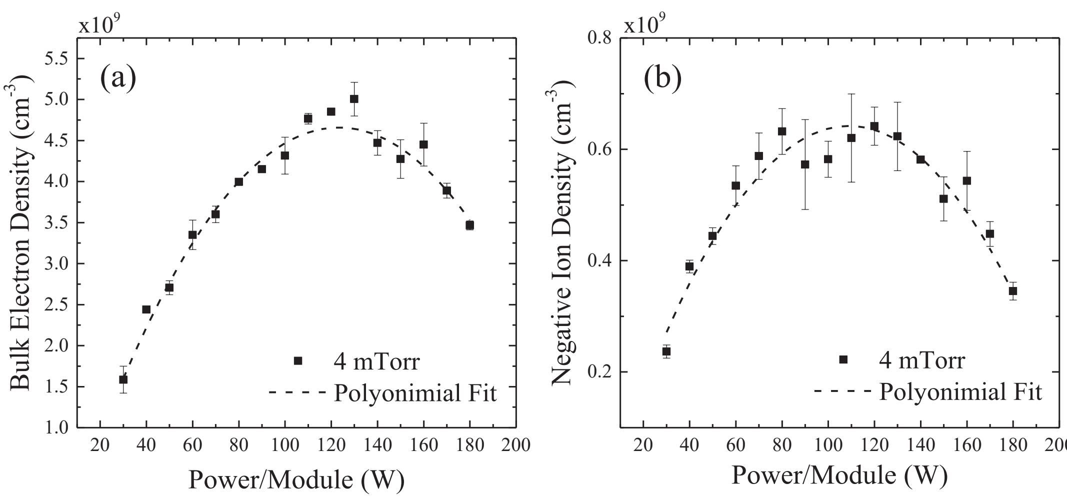

FIGURE 5. Power dependence of the (a) cold electron density and (b) negative ion density.

Negative ion H^- density versus power (Fig. 5b) exhibits similar to the cold electron density evolution, contrary the Lyman band emission (i.e. $B^1\Sigma_u^+$ excitation) increase in Fig. 4a. The density of negative ions depends on the balance between the production and destruction mechanisms. As regards the latter, there are three reactions to be taken into account: i) mutual neutralization in collisions with positive ions, ii) associative detachment in collisions with atoms, and iii) electron detachment[14]. Due to the low loss frequency of electron detachment, H^- losses should be dominated by processes i) and ii). It is unlikely that mutual neutralization is responsible for the reduction of negative ions, since the positive ion density (and thus the reaction rate) evolves similarly to the plasma density (Fig. 5a). Finally, as regards associative detachment, the reaction rate increases versus power since dissociation degree increases[12]. Thus, indeed, process ii) could partially be responsible for the H^- density profile in Fig. 5b. On the top of that, since the production of negative ions through DA is directly related to the low energy electron density, the agreement between the curve profiles in Figs. 5a and 5b indicates that the formation of H^- ions is limited by the cold electrons.

Furthermore, Fig. 6. shows that, as the power increases, not only the density of the cold electrons goes down (see EEDF peak) but at the same time their temperature goes up (see inset in Fig. 6), both resulting in reduced DA rate.

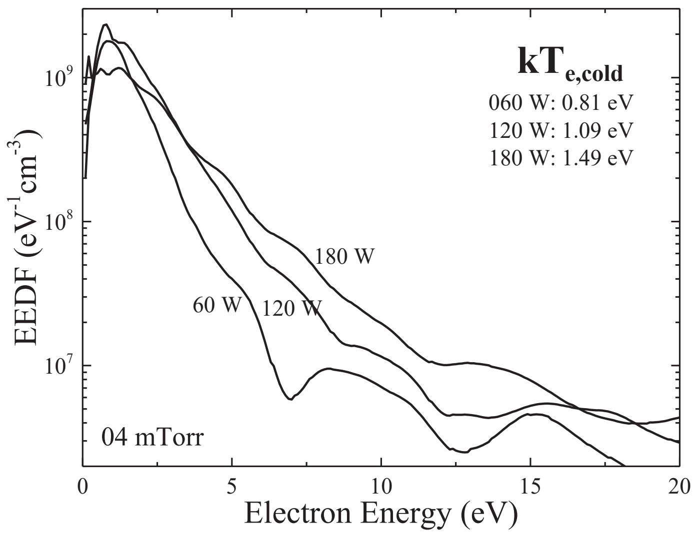

FIGURE 6. Electron energy distribution function for three indicative power levels.
$kT_{e,cold}$ refers to the energy of the cold electrons.

The Lyman-α intensity has an almost linear dependence on power (Fig. 7a). By eliminating the power variable (30–180 W/module) (Figs. 5b and 7a), the dependence of the negative ion density on the Lyman-α intensity (i.e. the peak of the curve obtained after Werner band subtraction) is experimentally obtained, and plotted in Fig. 7b.

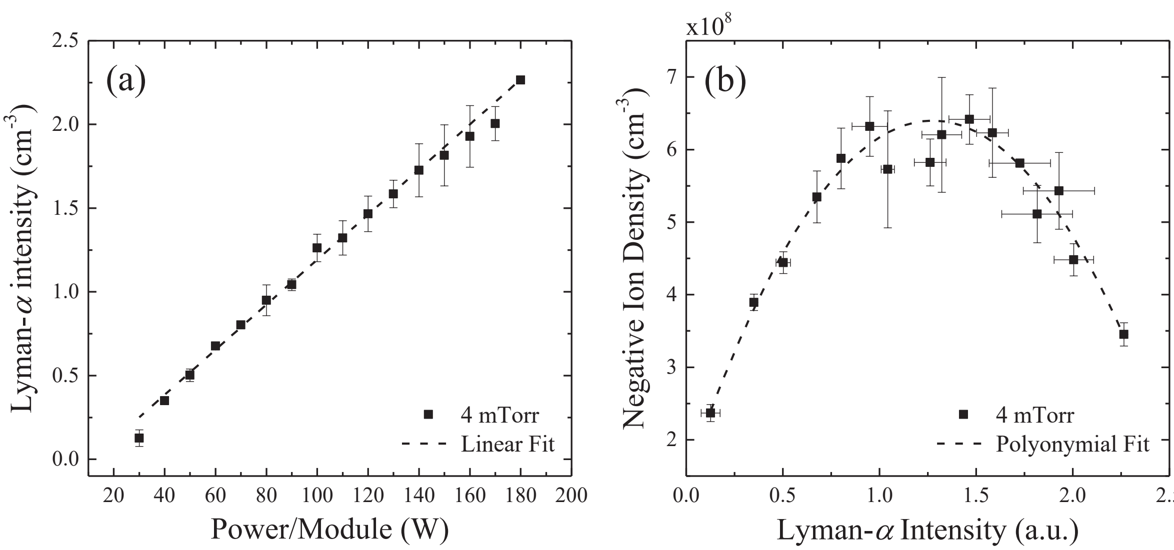

FIGURE 7. (a) Power dependence of Lyman-α intensity. (b) Negative ion density versus Lyman-α emission intensity.

Under the present specific conditions, a quadratic relation is not observed[24]. This fact implies that neutral resonant ionization does not significantly contribute to H^- formation in the present source. However, based solely on the present data, it is impossible to make the distinction among H^- fractions produced through different mechanisms. Such a distinction would require VUV absorption diagnostics for determining vibrational distributions and thus quantifying the absolute contribution of DA. Of similar importance would be the development of a self-consistent kinetic model[27], since it could provide all the parameters that are experimentally determined here, including the VUV-emission which is often neglected when modeling hydrogen plasmas and the vibrational distributions over wide range of working parameters.

CONCLUSIONS

VUV spectroscopy was conducted in the negative ion H^- source Prometheus I. Study of characteristic emissions in the VUV spectral range, in combination with electrostatic probe and laser photodetachment measurements, reasoned on possible mechanisms that create negative ions, by considering main destruction processes in parallel. The relation between the Lyman-α intensity and negative ion density was experimentally determined, due to its relevance to the production of negative ions through neutral resonant ionization. However, a quadratic relation between the above values, which would indicate the dominance of this mechanism, was not confirmed.

On the other hand, it appeared that the production of negative ions via dissociative attachment of cold electrons to vibrationally excited ground state molecules was consistent with the results. In the light of the available data, although associative detachment might impose a limitation to the H^- density, the volumetric rate of negative ion production can potentially be increased by providing more cold electrons (the source was found to be rich in vibrationally excited molecules).

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