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# Experimental Results for H<sub>2</sub> Formation from H<sup>-</sup> and H and Implications for First Star Formation

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**Abstract.** We have performed the first energy-resolved measurement of the associative detachment (AD) reaction  $\text{H}^- + \text{H} \rightarrow \text{H}_2^- \rightarrow \text{H}_2 + \text{e}^-$ , which is the dominant process forming molecular hydrogen in the early universe. We find excellent agreement between our measurements and the most recent theoretical calculation of the AD process. We also present model calculations of the formation of protogalaxies in the early universe utilizing the experimentally confirmed rate coefficient.

**Keywords:** Astrochemistry, Early Universe, Associative Detachment, First Stars

**PACS:** 34.50.Lf, 97.10.Bt, 97.20.Wt

## INTRODUCTION

According to the standard model the universe started out as a hot and homogeneous melange of matter and radiation. The cosmic microwave background that we observe today at a temperature of 2.7 K allows us to take a glance back in time to the era of recombination (redshift  $z \sim 1100$ ) when the universe still displays a remarkable uniformity with density fluctuations on the order of  $10^{-5}$  [1]. How these small perturbations could grow to form gas clouds which would eventually collapse gravitationally into galaxies and stars is not well understood [2].

This lack of understanding is noteworthy since primordial chemistry exhibits only a small fraction of the complexity of the present day interstellar medium. Only hydrogen, helium (relative abundance by mass 1:0.24) and traces of lithium and beryllium existed in the early universe, as all heavier elements were synthesized in stars and supernovae.

In order for a primordial clump of gas to collapse, radiative cooling through rovibrational transitions of molecular hydrogen is required, since cooling due to photon emission from excited H atoms becomes increasingly inefficient at temperatures  $T < 10000$  K [3, 4]. Without H<sub>2</sub> cooling the gas pressure would counteract the gravitational force and effectively bring the collapse to a halt.

In today's molecular clouds, the formation of H<sub>2</sub> is thought to proceed almost exclusively on the surface of dust grains, since the association of two neutral H atoms in the gas phase is very ineffective [5]. Molecular hydrogen formation in the early universe must have occurred in the gas phase, however, and the dominant pathway at redshifts relevant to first star formation is initiated by the formation of H<sup>-</sup>



which is followed by the associative detachment (AD) reaction with atomic hydrogen



While the reaction rate for the first step of this route – H<sup>-</sup> formation via radiative association of an H atom and an electron (Eq. 1) – is known with high precision [6], no consensus has been reached yet for the AD reaction (Eq. 2).

Three flowing afterglow studies of the AD reaction have been carried out in the past 40+ years [7, 8, 9]. All three measurements agree within their error bars. These measurements were all performed at room temperature and hence

they do not give any information on the energy dependence. Due to its relevance for early universe chemistry, the AD process has been calculated several times by different groups (see [10] and [11], and the supporting material on *Science Online* for a summary of previous experimental and theoretical results), however, the results disagree for the absolute magnitude of the rate coefficient, as well as for the general trend in the relevant temperature range (10 K-10000 K). Glover and Savin have shown that the uncertainty for this reaction is severely limiting our ability to make predictions for the collapse of protogalaxies in the early universe [10] and that new laboratory data are needed to resolve this issue.

## MEASUREMENT OF THE H<sub>2</sub> ASSOCIATIVE DETACHMENT RATE COEFFICIENT

A novel merged beams apparatus to study anion-neutral reactions was used to determine the rate coefficient for H<sub>2</sub> associative detachment. A detailed description of the apparatus has been published elsewhere [12]. A comprehensive analysis of the AD experiment and its uncertainties is given in [13]. Here, only a brief summary is given.

A strong H<sup>-</sup> beam is created in a duoplasmatron ion source (Peabody Scientific) and sent through a Wien filter which removes contaminants like O<sup>-</sup> and electrons. The first leg of the setup is used to collimate and steer the beam with appropriate ion optical elements before it is bent 90 degrees by a spherical electrostatic deflector, preventing direct line of sight from the ion source to the interaction and detection regions for background reduction. After some additional ion optics and diagnostics, the central part of the second leg serves as a photodetachment (PD) section. Here, the ion beam is superimposed with 1.4 kW of continuous wave infrared laser light at 975 nm, neutralizing ~10% of the H<sup>-</sup> beam by photodetachment

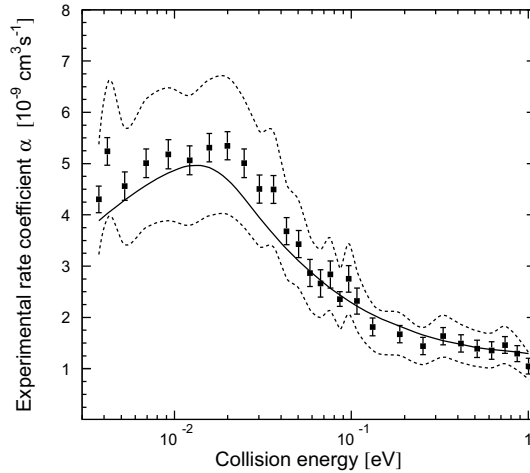


One of the main advantages of using a laser to produce the neutral H atom beam over traditional microwave or thermal disruption methods is that photodetachment will produce exclusively ground state H atoms, removing a significant source of uncertainty. To achieve a high fraction of neutralization, the output of an array of 19 × 24 individual laser diode emitters arranged in two interlaced stacks is bundled into a single beam with a focus of 4 mm × 7 mm (FWHM), that is nearly Gaussian in shape. Furthermore, a small intersection angle of 2.7 degrees between the ion beam and the laser was chosen to increase the overlap length and thereby enhance the PD efficiency. The center of the 2 m PD chamber houses a 1.2 m drift tube that can be biased with an independent negative voltage  $V_{\text{float}}$ . The photodetachment takes place well inside this drift tube whose purpose it is to create a variable velocity offset between the neutrals and ions that emerge from the PD section. While the H<sup>-</sup> ions are decelerated upon entering the drift tube, and accelerated back to their original velocity  $v_{\text{H}^-}$  at the end of it, the neutral H atoms that are created inside the drift tube region stay at the decreased velocity  $v_{\text{H}}$ . For the present experiment voltages  $V_{\text{float}}$  between -1 V and -281 V were used, resulting in H<sup>-</sup> on H relative collision energies  $E_{\text{coll}}$  in the co-moving center of mass frame ranging from 4 meV to 1 eV. The lower end of the collision energy scale is limited by the experimental resolution which is dominated by the angular spread of the merged beams that are confined by two 5 mm apertures before and after the PD region, 2.8 m apart.

In the interaction section the neutral and ion beams can either be merged to initiate the H<sub>2</sub> formation or switched on and off individually. The ion beam can be chopped by a fast (<100 ns) high voltage deflector plate at the entrance of the section while the neutrals can be chopped by switching the photodetachment laser with a 20 μs time constant. By means of a 100 Hz chopping scheme, background from H<sub>2</sub> molecules formed already inside the PD region as well as from other neutral- or ion beam related processes can be efficiently subtracted [12, 13].

Two rotating wire beam profile monitors are used to record the neutral and ion beam profiles during the measurement, which are needed to determine the merged beams overlap form factor  $\Omega$  [15, 16]. Behind the interaction region an electrostatic quadrupole deflects the H<sup>-</sup> ions into a Faraday cup that is used to continuously record the current. Typical values observed for a collimated H<sup>-</sup> beam are on the order of  $I_{\text{H}^-} = 650$  nA. The neutral current in the interaction section was determined by measuring the secondary electron emission in a calibrated “neutral” cup at the end of the beamline. On the order of 100 s<sup>-1</sup> H<sub>2</sub> molecules are created by the AD process in the interaction section and they have to be filtered out of a stream of ~10<sup>11</sup> neutral H atoms traveling alongside with the same velocity.

Our detection scheme makes use of the fact that the H<sub>2</sub> molecules possess twice the energy (20 keV) of the H atoms (10 keV). A differentially pumped gas cell is used to electron-strip a well-defined fraction of the neutrals and convert them into H<sub>2</sub><sup>+</sup>. In the gas cell chamber, we continuously bleed in helium, establishing a pressure of 2 × 10<sup>-4</sup> torr in an ~80 cm long section. For ~5 % of the H<sub>2</sub> particles passing this cell an electron is stripped off, leading to the formation of 20 keV H<sub>2</sub><sup>+</sup> ions. The H<sub>2</sub><sup>+</sup> particles are then filtered out from the flux of neutral (mainly H) and charged particles (mainly protons) in the next chamber by two consecutive cylindrical energy analyzers [17] and directed towards a



**FIGURE 1.** Rate coefficient for the associative detachment reaction  $\text{H}^- + \text{H} \rightarrow \text{H}_2 + e^-$ . The error bars mark the statistical error of the experiment while the dashed error bands show the uncertainty for the absolute scale. The solid line depicts the calculation of Cizek et al. [14] convolved with the experimental resolution.

channel electron multiplier (CEM) detector. The detector has a near-unity detection efficiency of  $(98 \pm 2)\%$  for  $\text{H}_2^+$  at these energies. After background subtraction, typical  $\text{H}_2^+$  rates  $R_{\text{H}_2^+}$  on the order of a few Hz are observed.

The helium column density  $N_{\text{He}}$  in the gas cell has been determined in an independent measurement observing the  $\text{H}^-$  loss by electron stripping in the gas cell [12]. The cross section for stripping of  $\text{H}_2$  in helium  $\sigma_{\text{strip}} = 1.044 \times 10^{-16} \text{ cm}^2$  is known with an accuracy of 16.5% [18]. Together with the uncertainty imposed by the unknown rovibrational state of the  $\text{H}_2$  molecules, prior to the stripping process, it constitutes the largest contribution to the uncertainty of our absolute scale.

Finally, the  $\text{H}_2$  AD rate coefficient is calculated by the formula

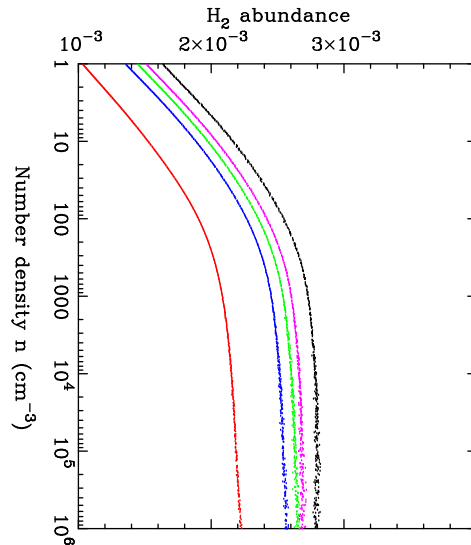
$$\alpha = \frac{R_{\text{H}_2^+}}{\sigma_{\text{strip}} N_{\text{He}}} \frac{e^2 v_{\text{H}^-} v_{\text{H}}}{I_{\text{H}^-} I_{\text{H}}} \frac{1}{\Omega}. \quad (4)$$

The rate coefficient as a function of the energy is shown in Figure 1. The systematic  $\pm 25\%$  uncertainty of the absolute scale is indicated with error bands. Also plotted is the result of the cross section calculation by Čížek *et al.* [14] that has been convolved into a rate coefficient with our experimental resolution. As can be seen, the agreement in shape and absolute scale is quite remarkable. The calculated values lie inside the experimental uncertainty over the entire energy range and hence we use the thermal rate coefficient  $\alpha_{\text{T}}$  calculated from the cross section given by Čížek *et al.* [14] (and extended to somewhat higher and lower energies [11]) as recommended reference.

## SIMULATION OF EARLY UNIVERSE PROTOGALAXY FORMATION

We have performed high-resolution simulations of primordial gas collapsing in a protogalactic halo. The simulation represents a low-mass protogalaxy in a relic HII region that has been ionized, e.g., by a distant Population III star. For details on the simulation see [11] and references therein. In Figure 2 the  $\text{H}_2$  abundance is plotted as a function of the total number density, where each point corresponds to a spherical shell centered about the cloud center. The results of five different runs are shown. Run 1 (black) used the previous upper limit for the AD reaction, Run 2 (red) marks the previous lower limit. Run 3 (green) used our new experimentally confirmed thermal rate coefficient. Run 4 (magenta) and Run 5 (blue) represent the remaining  $\pm 25\%$  uncertainty of the thermal rate coefficient, inferred from the uncertainty of the absolute scale of the measurement.

The difference in the  $\text{H}_2$  abundance has major implications for the temperature of the collapsing gas. The mass of the forming Population III star(s) is closely related to the Jeans mass at the minimum temperature. While the previous uncertainties translated into a factor of  $\sim 20$  uncertainty in the predicted mass of the Population III stars that are formed, our new rate coefficient limits this uncertainty to a factor of  $\sim 2$  (see [11] for details).



**FIGURE 2.** Simulation of primordial gas in an initially ionized protogalactic halo. The plot shows the  $H_2$  abundance for five different runs, using different values for the  $H_2$  AD reaction (see text for details).

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