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 Scientifiuqe, Ecole Polytechnique, 91128 Palaiseau Cedex, France
(Received 2 January 1979)

H^- and D^- densities are measured in low-pressure plasmas by photodetachment to confirm and extend previous work done with probes. Measurements indicate no isotope dependence, with n_- rising in proportion to n_e^3 in the density range below $n_e = 10^{10}$ cm^{-3}. This functional dependence is consistent with H^- production by a nonlinear process such as dissociative attachment to excited hydrogen molecules and H^- loss by diffusion.

Previous work[1] has shown that the H^- density in a hydrogen plasma is about 100 times larger than the density computed from the cross sections then known for H^- production and loss. However these results were questioned because of the method of measuring H^- density by Langmuir probes. The purpose of the present work is to confirm by an alternative method the existence of large densities of H^- ions in hydrogen plasmas and to extend the measurements to a deuterium plasma. This subject is of interest because of the need for sources of D^- ions for neutral injection into controlled fusion devices. The method developed for measurement of H^- ion density by photodetachment will be of use in higher-density plasmas where probe measurements are not possible.

Photodetachment has previously been used to measure negative-ion densities in oxygen plasmas.[2] We applied this technique to the measurement of H^- density in plasmas, since it has several advantages over the alternatives. The interpretation of photodetachment signals can be verified by comparison with known cross sections and by mass analysis of extracted ions.

Photodetachment ($H^- + \text{photon} \rightarrow H + e$) in a plasma produces an increase in electron density n_e without an immediate increase in the positive-ion density n_+. The change in n_e can be determined by probes or by microwaves, depending upon the geometry and plasma density. A light pulse from a ruby laser is suitable for this purpose because the photon energy (1.8 eV) is sufficient to detach the electron (0.75 eV affinity) but is too low to interact with the plasma by other single-photon processes such as photoionization. The photodetachment cross section is near its maximum (4×10^{-17} cm^2) at this energy.

To assure that the photodetachment signal is proportional to the density of H^- and not of other negative ions such as OH^-, O^-, or O_2^- (for which the cross sections are one or two orders of magnitude lower), we have magnetically analyzed the negative ions extracted from the plasma and also have verified (Fig. 1) that our measurements agree with the theoretical photodetachment fraction ($\Delta n_-/n_-$) computed from the cross section (σ_{ph}) for the photodetachment of H^-, but not with that for other ions:

$$\frac{\Delta n_-}{n_-} = 1 - \exp\left(-\frac{\text{laser pulse energy}}{\text{area}}\,\frac{\sigma_{ph}}{h\nu}\right). \quad (1)$$

The apparatus used for the photodetachment measurement[3] consists of a ruby laser capable of delivering pulses of 1 J energy and 30 ns duration, a probe circuit measuring the relative increase in n_e due to the laser pulse, and a calibrated light monitor measuring the energy of each pulse. Various tests were applied to assure the authenticity of the photodetachment signals. Figure 1 shows that for laser pulses above 0.1 J the photodetachment fraction is essentially unity and we can determine the relative density of H^- from the change in the probe electron-current density:

$$n_-/n_e = \Delta n_e/n_e = \Delta j_{\text{probe}}/j_{\text{probe dc}}. \quad (2)$$

Because of large noise signals at higher laser energies, most of our photodetachment data were measured at laser pulse energies between 0.1 and 0.2 J. Whenever experimental conditions changed we repeated the test indicated by Fig. 1 in order

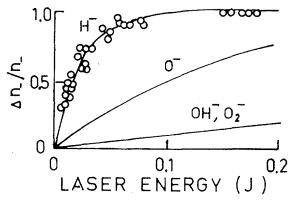

FIG. 1. Photodetachment of several negative-ion species by ruby laser light as a function of laser pulse energy, according to the theory of Eq. (1). Experimental data points are superimposed upon the theory of Eq. (1) for H^-, where $\sigma/h\nu = 139$ cm^2/J and the area of the laser beam is 3 cm^2.

to determine the appropriate range of laser pulse energies. The large signals at high laser power density (1 to 10 MW/cm^2) are due to the interaction of laser light with the probe surface and with surfaces at the plasma boundary. The probe technology is reported in Ref. 3.

Positive and negative ions were extracted from the plasma and mass analyzed by a magnetic mass spectrometer. The mass spectra measured with and without a cold trap indicate a strong effect of water vapor. The dominant species of negative and positive ions were H^- and H_3^+ when the trap was cold, but were OH^- and H_3O^+ when the trap was warm. Mass analysis confirmed that the photodetachment signal was correlated with the density of H^- ions and not with other species.

Figure 2 shows the dependence of n_- upon n_e in H_2 and D_2 plasmas confined only by their ambipolar potential.[4] The neutral-gas density was 2×10^{14} cm^{-3}. The proportion of hydrogen in the deuterium plasma did not exceed 10%. In H_2 the electron temperature varied in the range 0.10–0.40 eV, with a weak dependence upon n_e:

$$kT_e = (0.2\ \text{eV})\ln[n_e/(1.7\times10^8\ \text{cm}^{-3})]. \quad (3)$$

Figure 2 indicates that the negative-ion density is not isotope dependent and increases approximately in proportion to n_e^3 in the range of n_e between 1.7×10^9 and 1.1×10^{10} cm^{-3}. At maximum density the ratio n_-/n_e is 0.35.

Two mechanisms involving intermediate states were proposed[1] in order to explain the observed nonlinear dependence of the H^- density upon the plasma density and the large H^- density[5]: (1) dissociative attachment (DA) of electrons to vibrationally excited hydrogen molecules:

$$e + H_2(v) \rightarrow H_2^-(^2\Sigma_u^+) \rightarrow H(1s) + H^-; \quad (4)$$

(2) DA to an electronically excited long-lived state of hydrogen molecules:

$$e + H_2(^3\Pi_u) \rightarrow H_2^-(^2\Pi_u) \rightarrow H(2p) + H^-. \quad (5)$$

In 1978 theoretical[6] and experimental[7] work showed the cross sections for Reaction (4) to increase by four orders of magnitude from $v=0$ to $v=4$. The increase is even stronger for deuterium. Although DA to the ground state of the H_2 molecule is notorious for its strong isotope dependence, this dependence is rather weak for the higher vibrational levels, for which the cross section attains 4×10^{-16} cm^2. Estimates of the cross sections for Reaction (5) indicate[8] that they are in the range from 10^{-18} to 2×10^{-17} cm^2. No isotope dependence is expected in this case.

Although the recently acquired data[6-8] show that the DA cross section increases by orders of magnitude when H_2 is vibrationally or electronically excited, it is not yet possible to decide whether Reactions (4) and (5) can account for the H^- density observed in plasmas. Two peculiarities of these plasmas are at the origin of this difficulty:

(a) The neutral density is low (1–10 mTorr); therefore the unknown survival probability of vibrationally excited hydrogen molecules striking a wall is a critical parameter.

(b) The electron temperature is low ($\lesssim 1$ eV); therefore only the highest vibrational levels are effective in Reactions (4) and (5). If, however, Reaction (4) were responsible for the observed H^- density, then the principal source of hydrogen molecules in highly excited vibrational states has to be determined. Various possible production processes have been considered, such as charge exchange of H_2^+ and $H_2(v=0)$, wall neutralization of H_2^+, mutual neutralization, and electron collisions.[9] The contribution of dissociative recombination of H_3^+ ions, yielding hydrogen molecules in highly excited vibrational states, may also be important, but its rate is not yet known.

The functional form of the n_- dependence upon n_e^3 can be explained by a nonlinear H^- production process, such as DA to excited H_2, when the loss of H^- is dominated by diffusion. If we assume this production mechanism (cross section σ^*) to be dominating, the steady-state condition is

$$n_e n^* \langle \sigma^* v_e \rangle = n_-/\tau, \quad (6)$$

where n^* is the density of excited H_2.

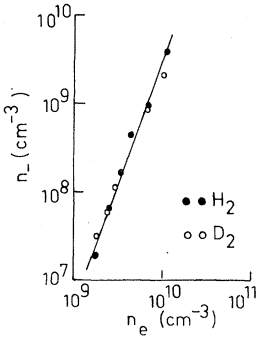

FIG. 2. Photodetachment measurements of the dependence of n^- upon n_e in H_2 and D_2. The ratio n_-/n_e was measured by a large probe valid for relative measurements. The value of n_e was measured separately by a Langmuir probe. The least-squares fit to the hydrogen data gives $n_- = 10^{-19} n_e^{2.85}$ with an uncertainty of 18%.

Since the excited molecules are produced by plasma collisions and destroyed by collisions with the wall, we should expect, for the low neutral and plasma densities used in this experiment, n^* to be proportional to n_e, for both vibrational and electronic excitation.

At low plasma density, when the H^- loss is dominated by diffusion, the lifetime can be shown to be proportional to n_e:

$$\tau_{\text{diff}} = n_e / C. \tag{7}$$

This follows from the following considerations[3,4]: The lifetime for diffusion of the H^- ions is determined by their flux across the potential barrier at the plasma surface; therefore it has a strong dependence upon the ratio of electron and negative ion temperatures,

$$\tau_{\text{diff}} = \frac{V}{S}\left(\frac{2\pi m_-}{kT_-}\right)^{1/2}\left[\left(\frac{m_+}{m_e}\right)^{1/2}\frac{n_e}{n_+}\right]^{T_e/T_-} \tag{8}$$

(V/S is the plasma volume to surface ratio). Equation (7) is compatible with (8) and with the empirical dependence (3) if $kT_- \simeq 0.4$ eV and $C = 1.6\times10^{13}$ cm^{-3} s^{-1}.

n_- is expected to rise in proportion to n_e^3 only up to densities when the loss of negative ions by destruction (mutual neutralization and detachment in electron collisions) becomes dominating. The transition to a new functional form of the n_- dependence upon n_e will occur when $\tau_{\text{destr}} = \tau_{\text{diff}}$, where

$$\tau_{\text{destr}}^{-1} = 2\times10^{-7} n_e. \tag{9}$$

For H^- the corresponding n_e is 1×10^{10} cm^{-3}, which is at the upper limit of our measurements. At higher densities, n_- is expected to increase only linearly with n_e. It is then possible to make a conservative extrapolation of this model to the densities required for controlled fusion injection ($n_- \geq 10^{12}$ cm^{-3}).

This project was performed at the Ecole Polytechnique as a joint project with Office National d'Etudes et Recherches Aérospatiales and Lawrence Livermore Laboratory, funded by the French Commissariat à l'Energie Atomique and the U. S. Department of Energy under Contract No. W-7405-Eng-48. Valuable consultations were provided by H. J. Doucet, J. R. Hiskes, and J. Taillet.

---

[a] Permanent address: Lawrence Livermore Laboratory, Livermore, Calif. 94550.

[1] E. Nicolopoulos, M. Bacal, and H. J. Doucet, J. Phys. (Paris) 38, 1399 (1977), and in Proceedings of the Symposium on Production and Neutralization of Negative Hydrogen Ions and Beams, Brookhaven National Laboratory, 26-30 September 1977, edited by K. Prelec, BNL Report No. 50727 (unpublished).

[2] J. Taillet, C. R. Acad. Sci., Ser. B 269, 52 (1969).

[3] M. Bacal, G. W. Hamilton, A. M. Bruneteau, H. J. Doucet, and J. Taillet, to be published, and Laboratoire de Physique des Milieux Ionisés Report No. PMI 896, 1978 (unpublished).

[4] M. Bacal and H. J. Doucet, IEEE Trans. Plasma Sci. 1, 91 (1973).

[5] Experimental work by B. Peart, R. A. Forrest, and T. K. Dolder (to be published) has shown that the cross section for the reaction $e + H_3^+ \rightarrow H_2^+ + H^-$ never exceeds 2×10^{-18} cm^{-3} and the electron threshold energy is approximately 1.5 eV. This cross section is less than that for $e + H_2^+ \rightarrow H^+ + H^-$, considered in Ref. 1. Therefore dissociative recombination of H_3^+ cannot account for H^- production in plasma.

[6] J. M. Wadehra and J. N. Bardsley, Phys. Rev. Lett. 41, 1795 (1978).

[7] M. Allan and S. F. Wong, Phys. Rev. Lett. 41, 1791 (1978).

[8] B. D. Buckley and C. Bottcher, private communication.

[9] J. R. Hiskes, M. Bacal, and G. W. Hamilton, Lawrence Livermore Laboratory Report No. UCID-18031, 1979 (unpublished).