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Neutral hydrogen in high-temperature pinch plasmas and its influence on the ionization dynamics of impurities

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The neutral-hydrogen density in a hot pinch plasma and in the surrounding halo is investigated spectroscopically, and its influence on the ionization dynamics of impurity ions is analyzed. The results are employed in the analysis of effective-ionization-rate coefficients, which are derived for Si VIII to Si XII from the time evolution of the respective ions in the plasma.

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Spectroscopic observations of time-dependent hot plasmas have been used by several groups to derive effective rate coefficients for ionization, recombination, and excitation of multiply ionized atoms. In most cases, the atomic species of interest were added in small amounts to discharges in hydrogen. The plasmas had to be well diagnosed, preferably employing other diagnostic methods, and usually laser scattering was used. The general method has been discussed previously (for instance, Refs. [1] and [2]), and a review of all results has been carried out by Griem [3]. When this method is compared with techniques that measure cross sections employing crossed electron and ion beams, it certainly has the disadvantage that it will not yield cross sections with all their detailed structures for comparison with theoretical calculations. On the other hand, the "plasma spectroscopic method" yields *effective* rate coefficients, which are ultimately needed in most applications; details of the cross sections are averaged, but possible additional channels are included, whose contributions are sometimes difficult to assess.

Rate coefficients for ionization are best derived from plasmas in which the respective ions are in a rapidly ionizing regime. Pinch experiments and also sawtooth oscillations of tokamak plasmas have been used. As the review by Griem [3] reveals, most measurements were carried out on Θ-pinch discharges, and the agreement of the rate coefficients with theory was excellent for hydrogenlike ions and acceptable for heliumlike ions. However, for low-Z lithiumlike ions as well as for low charge states of other ionic species, deviations between experiment and theory were large, the experimental values being, on the average, only 60% of the theoretical ones.

This trend toward small effective rate coefficients for ionization from plasma measurements was observed by several groups employing different Θ-pinch devices (e.g., Brown *et al.* [4], Kunze [5], and Greve *et al.* [6]). Since in the meantime cross-section measurements have confirmed the theoretical values in many cases, possible sources for this effect in pinch discharges have been sought. Charge exchange with neutral hydrogen is one possibility.

The densities n_z of the ionization stages z of an atomic species in a plasma of electron density n_e are governed by the following set of rate equations:

$$\frac{\partial n_z}{\partial t}=n_e(n_{z-1}S_{z-1}-n_zS_z)+n_e(n_{z+1}\alpha_{z+1}-n_z\alpha_z)-\nabla\cdot\Gamma_z\ , \qquad (1)$$

where S_z is the total effective rate coefficient for ionization, α_z is the effective rate coefficient for recombination, and Γ_z represents the particle flux. For ions in a rapidly ionizing regime, which holds for most pinch discharges, recombination usually is negligibly small, and the time evolution of the ionization stages is essentially determined only by the ionization rates,

$$\frac{\partial n_z}{\partial t}=n_en_z\left[\frac{n_{z-1}}{n_z}S_{z-1}-S_z\right]\ , \qquad (2)$$

which therefore can be derived from observations.

If charge exchange (cx) is important, it has to be taken into account by adding its contribution to Eq. (2):

$$\left.\frac{\partial n_z}{\partial t}\right|_{\mathrm{cx}}=-n_{\mathrm{H}}n_z\langle v\sigma_z^{\mathrm{cx}}\rangle+n_{\mathrm{H}}n_{z+1}\langle v\sigma_{z+1}^{\mathrm{cx}}\rangle\ . \qquad (3)$$

$\langle v\sigma_z^{\mathrm{cx}}\rangle$ is the effective rate coefficient. Since the atomic species are introduced into the hydrogen discharges usually in amounts of less than 1%, the electron density equals the proton density, $n_e=n_{\mathrm{H}+}$. The net effect of charge exchange is to slow down ionization, and the effective-ionization-rate coefficients of Eq. (2) become

$$\begin{aligned}S_{z,\mathrm{eff}}&=S_z\left[1-\frac{n_{z+1}}{n_z}\frac{n_{\mathrm{H}}}{n_{\mathrm{H}+}}\frac{\langle v\sigma_{z+1}^{\mathrm{cx}}\rangle}{S_z}\right]\\&=S_z\left[1-\frac{n_{z+1}}{n_z}s_z\right]\ . \qquad (4)\end{aligned}$$

It is obvious that the magnitude of the relative neutral hydrogen concentration $n_{\mathrm{H}}/n_{\mathrm{H}+}$ will determine the importance of this effect and that it will most strongly influence the tail of any transient ionization stage, where $n_{z+1}/n_z\gg 1$.

Two observations indeed pointed to the possibility of charge exchange in hydrogen pinch plasmas. When studying recombination into higher levels of lithiumlike ions, Datla and Kunze [7] found effective rates that were much larger than all known theoretical rates for radiative, three-body, and dielectronic recombination. They concluded that charge exchange could explain the observations if the neutral hydrogen density was higher than theoretically estimated by at least a factor of 100. Schmidt *et al.* [8] measured rate coefficients for ionization of Ne VI to Ne VIII in a helium instead of a hydrogen discharge and found good agreement with theory. Using theoretical estimates for the densities of neutral hydrogen and neutral and ionized helium, respectively, they concluded at that time that in neither of the discharges should charge exchange be important. In order to shed light on these problems, we measured the hydrogen concentration in our pinch discharge.

The Θ-pinch device has been described in detail previously [6]. The typical initial filling pressure is 1.6 Pa, and with a reversed-bias magnetic field, a stable plasma column is produced; its length is about 18 cm and its diameter is 4 cm. Electron density and temperature as a function of time are obtained along a diameter of the plasma column in the midplane of the discharge using Thomson scattering. At the time of maximum current (5.75 μs after initiation of the main discharge) the electron density reaches 3×10^{16} cm^{-3} and the electron temperature 235 eV. The ion temperature of impurity species is deduced from Doppler profiles of respective emission lines and is about 900 eV. This value is also obtained from pressure-balance considerations for the temperature of the protons.

We now measured intensities and profiles of the H_α and H_β lines in the midplane of the discharge tube using a 1-m monochromator equipped with an RCA-4526 photomultiplier. A mirror system allowed observation along any chord of the cross section, and *local* emission coefficients were thus derived through Abel inversion. The complete system was calibrated absolutely employing a tungsten strip lamp.

The profile of the H_α line was scanned recording the emission along a diameter from discharge to discharge. The shape was well approximated by a Gaussian function, and taking into account the apparatus function, a hydrogen temperature of (1.6±0.4) eV was derived. It represents an average temperature along the diameter of the discharge tube, and it remained practically constant during the lifetime of the plasma. One should keep in mind that this temperature is an upper limit since, for the magnetic fields of the discharge (up to 2.5 T at current maximum), the Zeeman effect is of the same magnitude as Doppler broadening [9]. The observation was perpendicular to the magnetic field.

The observed hydrogen is rather cold compared with the ion temperature of the plasma and, as will be shown later, the emission originated from hydrogen outside the hot plasma column and its boundary. The width of the H_β line was also narrow and indicated that the electron density in the region of the cold hydrogen was of the order of 10^{14} cm^{-3} or even lower. One should keep in mind that any H_α emission from hot hydrogen atoms representative of the plasma temperature would have a very broad spectral profile, and the contribution to the emission would be barely discernable from the continuous background.

For the intensity measurements the width of the exit slit of the monochromator was chosen such that the total line was recorded. Figure 1 shows as an example the radiance L of the H_α line as a function of the distance from the axis as measured in the midplane of the plasma column at 6 μs after initiation of the discharge. The local emission coefficient ϵ as a function of radius was obtained by Abel inversion. The maximum of the emission is at a radius of $r\approx3$ cm, i.e., just outside the hot plasma column, whose boundary from Thomson scattering is at $r=2.2$–2.5 cm [6]. The wall of the discharge tube is at $r=10$ cm.

The time development of the hydrogen emission is shown in Fig. 2. The emission coefficient ϵ of the H_α line is displayed as a function of radius for various times during the discharge. At early times, the emission is strong at the wall. During the discharge, it decreases at the wall and the maximum at the boundary of the hot plasma develops. The H_β emission matches the H_α emission.

The emission coefficient ϵ of a line $(p\rightarrow q)$ is given by

$$\epsilon(p\rightarrow q)=\frac{h\nu}{4\pi}A(p\rightarrow q)n(p)\ , \tag{5}$$

where $h\nu$ is the energy of the photons and $A(p\rightarrow q)$ is the transition probability. From the absolute values of the emission coefficients of H_α and H_β the population densities of hydrogen in the levels with the principal quantum numbers 3 and 4 can be calculated. For the plasma condition at $t=6$ μs, for example, we obtain

$$n_H(3)=4.6\times10^{8}\ \text{cm}^{-3},\quad n_H(4)\approx3.9\times10^{8}\ \text{cm}^{-3}\ ,\qquad r=3\ \text{cm}$$

$$n_H(3)=1.5\times10^{8}\ \text{cm}^{-3},\quad n_H(4)\approx1.3\times10^{8}\ \text{cm}^{-3},\qquad r=1.5\ \text{cm}\ .$$

Electron transfer from hydrogen atoms in excited

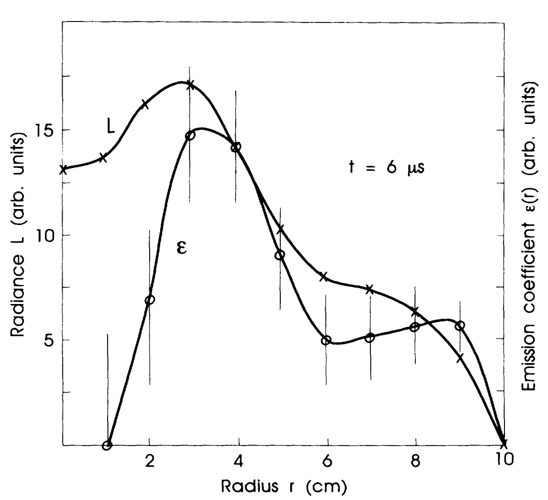

FIG. 1. Spectral radiance and emission coefficient of the H_α line in the midplane of the plasma column at $t=6$ μs.

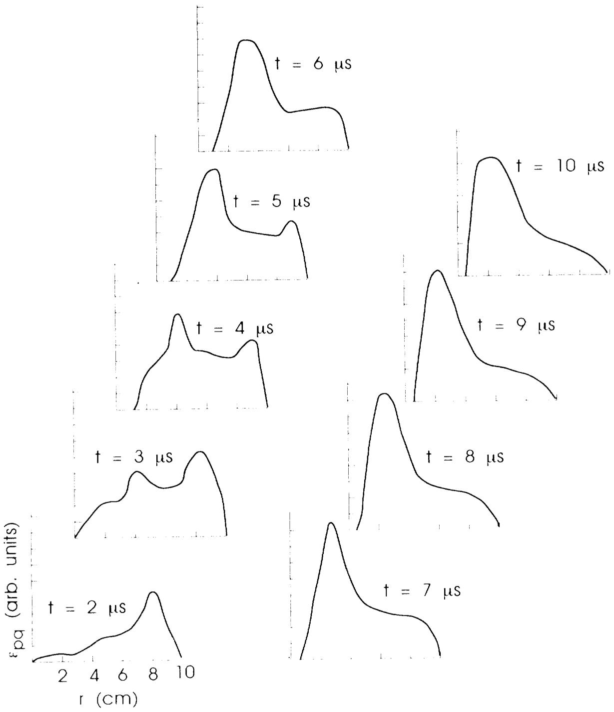

FIG. 2. Time evolution of the H_α emission coefficient in the midplane of discharge.

states can make an important contribution, since the corresponding cross sections are large: they scale with the fourth power of the principal quantum number p_H of the respective state of the hydrogen atom [10]. On the other hand, the principal quantum number p_A of the ionic state, which is most probably occupied after the collision also increases, in fact linearly with p_H; since high-lying levels start to become collisionally coupled to the continuum with increasing plasma density, such capture processes may not result in effective recombination and therefore will not modify the ionization-rate coefficient. This quantum number of the most probably occupied state for collisions with hydrogen atoms is approximately given by [11]

$$p_A \approx p_H Z^{3/4} , \tag{6}$$

and it should be compared with the collision limit p_c of Griem [12]:

$$p_c \simeq 126 Z^{12/17} n_e^{-2/17} (kT_e/E_H)^{1/17} , \tag{7}$$

where the electron density n_e is in cm^{-3} and the electron temperature kT_e/E_H is in units of the Rydberg energy E_H. For $p_A > p_c$, electron capture from an excited state (p_H) leads to negligible recombination, and charge exchange with hydrogen atoms in their ground state will give the major contribution, if charge exchange is important at all. With $p_H = 1$, an approximate upper limit can thus be derived for the electron density, where charge exchange with neutral hydrogen may play a role in the ionization dynamics of impurity ions in a plasma. The combination of Eqs. (6) and (7) yields for this limit

$$n_e \leq (7 \times 10^{17}) Z^{-3/8} (kT_e/E_H)^{1/2} . \tag{8}$$

The densities of our pinch plasma are in that regime.

We now have to estimate the ground-state density of the hydrogen atoms from the measured population densities of the $p=3$ and 4 levels, and we start with the hot pinch plasma at $r = 1.5$ cm. The thermal limit p_{th}, which for plasmas in ionization balance gives the principal quantum number of the lowest level whose population density is in partial thermodynamic equilibrium with all higher levels and the continuum of free electrons, is for our conditions $p_{\text{th}} \approx 2$ (Ref. [12]). The population densities of the upper levels of the H_α and the H_β lines should be given therefore by the Saha-Boltzmann equation, and we obtain $n(3) = 7.5 \times 10^8$ cm^{-3} and $n(4) = 1.3 \times 10^9$ cm^{-3}. The experimental population densities are much lower than these values, and the explanation is obvious: our measurements recorded only that fraction of the hydrogen atoms that were cold and were entering the hot plasma from the outside. Their population densities are not in equilibrium, the atoms experience an *ionizing phase,* and their ground-state density $n(1)$ has to be derived accordingly. From both upper levels we obtain an average density of $n(1) \simeq 3 \times 10^{10}$ cm^{-3} using the calculations of Refs. [13] and [14].

It is nearly impossible to arrive at a reliable estimate of the hydrogen density outside the hot plasma column, since the plasma parameters of this plasma region—called the halo [15]—are poorly known. For the electron density we estimate the upper value from the experimental width of the H_β line of $n_e \approx 1 \times 10^{14}$ cm^{-3}, which is consistent with investigations on a smaller discharge tube at somewhat higher filling pressures [15]. Also, the electron temperature is rather uncertain. The relative intensities of the H_α and H_β lines correspond to an excitation temperature of 0.9 eV; the profile of the H_α line gives an upper limit on the temperature of the hydrogen atoms of 1.6 eV, which is also consistent with Ref. [15]. If we adopt this temperature for the electrons, the collisional-radiative model of Johnson and Hinnov [16] predicts a ground-state density of the hydrogen atoms of $n(1) \approx 5 \times 10^{13}$ cm^{-3} for the excited-state populations measured at $r = 3$ cm, assuming that the plasma is optically thick to Lyman lines. The calculations of Drawin and Emard [17] yield a somewhat higher value. However, these derivations are extremely sensitive to the electron temperature in this temperature region: a temperature of 2.8 eV instead of 1.6 eV would result in ground-state densities in the halo region that are *lower by more than a factor of 10.*

The influence of cold hydrogen atoms entering the hot plasma should also be seen on the emission lines of ions that are affected by charge exchange. The best candidates are $\Delta p = 1$ transitions between high-lying levels of multiply ionized atoms in the visible spectral region, and for that purpose we added small amounts of SiH_4 to the discharge (the Si density was less than 1% of the electron density) and studied the emission of the $p = 10 \rightarrow 9$ transition of Si IX at 479.6 nm and the $p = 12 \rightarrow 11$ transition of Si XII at 479.4 nm. Both ionization stages exist during different times of the discharge. The emission of both

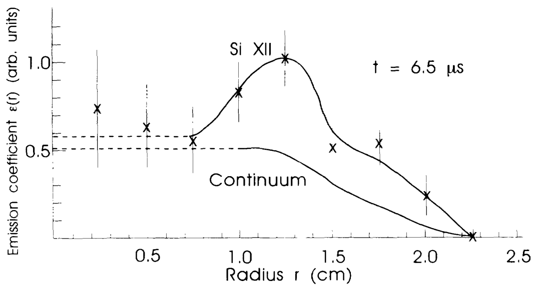

FIG. 3. Radial distribution of the emission coefficients of the continuum and of the $p=12\rightarrow 11$ transition of Si XII at $t=6.5$ μs.

lines displayed similar behavior, and Fig. 3 shows as an example the radial distribution of the emission coefficient of the Si XII line at $t=6.5$ μs. The error bars are large in the central region, a consequence of the Abel inversion. Also shown is the continuum emission. Both $\Delta p=1$ lines display enhanced emission towards the boundary with two bumps at $r\approx 1.25$ cm and $r\approx 1.75-2$ cm. This is indeed evidence of charge exchange of Si ions with the cold hydrogen atoms, the density of which increases by about several orders of magnitude from the hot plasma to the halo region.

There is certainly no influence of charge exchange on ionization in the central plasma region. It is possible, however, when the hot impurity ions are reflected at the plasma boundary with the high influx of cold hydrogen. For an estimate we look at the ionization correction factor s_z introduced in Eq. (4),

$$s_z=\frac{n_{\rm H}}{n_{{\rm H}^+}}\frac{\langle v\sigma_{z+1}^{\rm cx}\rangle}{S_z} . \tag{9}$$

Recent results revealed that electron-capture cross sections in collisions with H atoms are also large in low-energy collisions [18], the cross sections being of the order of $\sigma\approx 10^{-14}$ cm^2. The thermal velocity of Si ions at a temperature of 900 eV is 5.6×10^6 cm s^{-1}, yielding $\langle v\sigma_{z+1}^{\rm cx}\rangle\approx 6\times 10^{-8}$ cm^3 s^{-1}. If we take the ionization-rate coefficient of Si X as an example, $S_z\approx 5\times 10^{-11}$ cm$^3 s^{-1}$, and $n_{{\rm H}^+}=n_e$, we obtain

$$s_z\approx 10^3\frac{n_{\rm H}}{n_e} .$$

This shows that one needs $n_{\rm H}/n_e\geq 10^{-4}$ for charge exchange to influence ionization ($s_z\geq 0.1$). This condition is not fulfilled in our plasma at $r=1.5$ cm, where we have $n_{\rm H}/n_e\approx 10^{-6}$ for the cold hydrogen atoms. It is fulfilled in the boundary region, but in order to influence the overall ionization averaged over the total plasma volume, the neutral hydrogen density must be much larger.

We conclude, therefore, that for our present plasma conditions an influence of ionization by charge exchange is rather unlikely. However, it is quite conceivable that this is the case for plasmas with higher neutral hydrogen density in the halo region. This can easily occur for apparently the same discharge conditions, if the preionization discharges change slightly and the pickup of the initial plasma during the first implosion of the pinch discharge is less effective.

We derived effective-ionization-rate coefficients for Si VIII to Si XII from the time histories of suitable lines following the procedures as discussed in Refs. [1]–[6]. The Si concentration was 0.6%, and all lines were optically thin at the ion temperature of 900 eV. This time, the length of the plasma column was also measured from the emission of high-lying $\Delta p=1$ lines of Si ions as function of time, in contrast to the previous experiments [19], where it was obtained from the continuum emission. All ions have either a non-negligibly populated low-lying level or a metastable level (m), from which ionization contributes. We introduce the ratio $\beta=n_z(m)/n_z(g)$ of the population densities, where (g) refers to the ground state, and obtain the effective-ionization-rate coefficient [2]

$$S_z^{\rm eff}=\frac{1}{1+\beta}S_z(g)\left[1+\beta\frac{S_z(m)}{S_z(g)}\right]=\rho S_z(g) . \tag{10}$$

β is determined experimentally for Si IX to Si XII [20], and the ratio of the ionization-rate coefficients, $S_z(m)/S_z(g)$, is derived employing the semiempirical formula of Lotz [21].

Table I gives the experimental effective-ionization-rate coefficients $S_z^{\rm eff}$ together with the electron temperature at the peak emission of the respective ion, the factor ρ, and the ground-state ionization-rate coefficient $S_z^{\rm expt}(g)$ calculated according to Eq. (10). For Si VIII, ρ is taken to be 1 since no measurements of population densities of excited states were available. The column $S_z^{\rm Lotz}(g)$ shows the semiempirical rate coefficients of Lotz [21], and the columns $S_z^{\rm Arn}(g)$ and $S_z^{\rm Len}(g)$ those from the compilations of Arnaud and Rothenflug [22] and Lennon *et al.* [23], respectively. Ratios of experimental values and those of Ref. [23] are shown in the last column to illustrate the agreement. This is quite satisfactory, especially if one keeps in mind that errors of 10% in density and in temperature result in errors of 10% and 25–30 % in the rate

TABLE I. Comparison of experimental and theoretical ionization-rate coefficients. Numbers in square brackets denote powers of 10.

Ion	kT_e	$S_z^{\rm eff}$	ρ	$S_z^{\rm expt}(g)$	$S_z^{\rm Lotz}(g)$	$S_z^{\rm Arn}(g)$	$S_z^{\rm Len}(g)$	$S_z^{\rm expt}/S_z^{\rm Len}$
Si VIII	195	2.3 [−10]	≈1	2.3 [−10]	2.9 [−10]	3.3 [−10]	2.5 [−10]	0.9
Si IX	230	1.4 [−10]	≈1	1.4 [−10]	1.9 [−10]	2.2 [−10]	1.8 [−10]	0.8
Si X	235	8.4 [−11]	1.05	8.0 [−11]	9.5 [−11]	1.0 [−10]	9.0 [−11]	0.9
Si XI	225	4.3 [−11]	1.04	4.1 [−11]	3.5 [−11]	3.2 [−11]	3.1 [−11]	1.3
Si XII	215	1.4 [−11]	1.03	1.3 [−11]	1.1[−11]	1.0 [−11]	9.7 [−12]	1.3

coefficients, respectively. Also, the simple Lotz formula [21] again provides good estimates of the rate coefficients.

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