Destruction of the 3s State in Atomic Hydrogen by Fast-Atom Impact on N_2, Ar, and H_2 †

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Cross sections have been determined for the destruction of the 3s state in atomic hydrogen by 10-, 25-, and 35-keV hydrogen-atom impact on N_2, Ar, and H_2. The experimental values support the concept of two destruction mechanisms. One mechanism involves the collisional ionization of the excited atom, which has been treated quantitatively by Bates and Walker. The other mechanism involves electron capture by the fast-atom nucleus.

INTRODUCTION

Bates and Walker[1] have treated the collisional ionization of fast excited hydrogen atoms passing through a neutral gas. One of the basic assumptions of their theory is that the field of the nucleus of the fast atom is important only in that it determines the orbital contribution to the electron velocity relative to the target atom and also determines the binding energy of the electron. They assumed that the energy required to set the electron free would be provided in the elastic collision between the electron and the target atom. Thus by neglecting the field of the nucleus, the problem is reduced to an electron-atom collision.

They developed a formula in which the experimentally determined total electron scattering cross sections[2] are used. They computed electron-loss cross sections for impact on N_2. At the $n=3$ level and for energies beyond 10 keV, the computed electron-loss cross section versus atom velocity curve lies close to the total electron cross section versus electron velocity curve. (The electron velocity approximates the fast-atom velocity in our energy range beyond 10 keV since the translational velocity of the center of mass is larger than the mean orbital velocity of the electrons.) This was the only quenching mechanism they considered.

APPARATUS AND PROCEDURE

Protons were passed through a differentially pumped 8-cm-long collision chamber. The partially neutralized beam passed through the exit aperture into an evacuated observation region. The beam was highly collimated so that no protons intercepted the edge of the $\frac{1}{16}$-in.-diameter exit aperture. An EMI 9558B photomultiplier fitted with an H_α interference filter detected the $3s\rightarrow 2p$ radiation at a distance of $v\tau$ from the exit aperture, where v is the atom velocity and τ is the lifetime of the 3s state (160 nsec). At this point we were assured that the shorter-lived 3d and 3p states had decayed to a negligible fraction of the light.

An expression has been developed which describes the buildup of fast excited atoms in such a collision chamber.[3] It can easily be modified to the form used in this experiment:

$$\frac{I}{n_+^0\rho}=K\left(\frac{\sigma_s/\sigma_c+Q_x/Q}{L/v\tau+\alpha}\,(1-e^{-L/v\tau-\alpha}) + \frac{(1-Q_x/Q)(e^{-\beta-\gamma}-e^{-L/v\tau-\alpha})}{L/v\tau-(\beta+\gamma)+\alpha}\right), \quad (1)$$

where I is the intensity of the $3s\rightarrow 2p$ radiation emanating from the beam; n_+^0 is the proton density in the beam at the entrance aperture of the collision chamber; ρ is the target gas density in the collision chamber; K is a constant at a given energy; L is the collision chamber length; σ_s the total stripping cross section; σ_c is the total electron-capture cross section; Q_x is the cross section for excitation of ground-state hydrogen atom to the 3s state by atom impact on the target gas; Q is the cross section for electron capture into the 3s state of hydrogen by proton impact; $\alpha=\rho L Q_i$, where Q_i is the cross section for collisional destruction of the 3s state; $\beta=\rho L\sigma_c$; and $\gamma=\rho L\sigma_s$.

The intensity of the $3s\rightarrow 2p$ light was measured as a function of the target gas pressure in the collision chamber. The proton density n_+^0 was determined by using a secondary electron detector. The secondary electron detector measured the total particle density in the beam since neutrals, produced by charge transfer in the collision chamber, and protons have nearly the same detection efficiency.[4] The gas density ρ was measured with a calibrated ion gauge. The quantities σ_s and σ_c were taken from Allison.[5] The cross section Q was obtained from Hughes *et al.*[6] and the cross section Q_x was taken from Hughes *et al.*[7]

Figure 1 shows a computer fit of Eq. (1) to data for impact on Ar using Q_i as the fitting parameter. In principle, two points on the $I/n_+^0\rho$ versus ρ curve is sufficient to make one determination of Q_i.

Table I displays our values for the destruction cross section Q_i along with values for the collision-

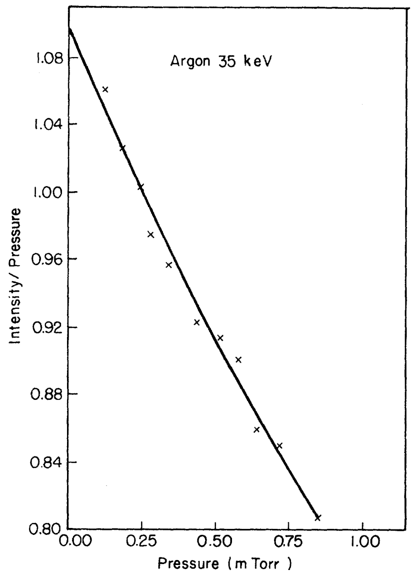

FIG. 1. Intensity per unit pressure of the $3s \rightarrow 2p$ radiation as a function of the collision chamber pressure. The target gas is argon. The line is a computer fit of Eq. (1) to the data points using Q_t as the fitting parameter.

al ionization cross section σ_i which are computed by the method of Bates and Walker. It is apparent that collisional ionization is not sufficient to explain the destruction of the $3s$ state, particularly by impact on H_2 and Ar. Therefore, another mechanism is postulated.

The basic assumption of the theoretical work of Bates and Walker is that the field of the proton can essentially be neglected so that the electron in the excited atom can be treated as a particle independent of the nucleus. Bates and Walker test this assumption by comparing their computed electron-loss cross sections for normal ($n = 1$) atom impact on various gases with experimental stripping cross sections. They show that their computation reproduces the general trend from gas to gas, and their computed values are greater than the experimental values for stripping by less than a factor of 2. Their computed values were expected to be too large because of the neglect of the screening of the effective charge of the electron by the proton. The screening, as they point out, is less important for excited hydrogen atoms and therefore their assumptions should be valid for collisions involving excited atoms.

These same arguments support the point of view that, conversely, the screening of the proton by the electron can be neglected in the case of the excited hydrogen atom. Thus the proton can also be treated as a separate particle in the same sense that Bates and Walker treat the electron as a separate particle. In this approximation it becomes clear that electron capture is an important mechanism that should not be neglected. Two mechanisms are in operation here

$$H^* + T \rightarrow H^+ + T + e, \tag{2}$$

$$H^* + T \rightarrow (H + e) + T^+ , \tag{3}$$

where H^* is the excited hydrogen-atom projectile and T is the target atom (or molecule). Reaction (2) is the collisional ionization mechanism as treated by Bates and Walker. Reaction (3) is the electron-capture reaction where the $(H + e)$ complex can be either a bound H^- or an unbound H^- system.

Figure 2 shows cross sections for ionization of the $n = 3$ atom which have been computed from the Bates and Walker formula using the total electron scattering cross sections from Ref. 2. Also shown are the sums of the ionization cross sections and the total electron-capture cross sections.[5,8] These sums should approximate the total destruction cross sections.

Table I presents a comparison of the measured destruction cross sections with the sum of the cross sections for ionization of the $n = 3$ atom and the total electron-capture cross sections. The uncertainties in Q_t are estimated uncertainties based on the reproducibility of the original data, the uncertainty in the ratio of Q_x/Q, and the uncertainty in ρ. The absolute calibration errors in determining Q_x and Q are not important in this experiment since Q_x and Q were measured in the same laboratory. The uncertainty in Q_x/Q was taken to be the sum of the reproducibility uncertainties in Q_x and Q. The some-

TABLE I. Experimental destruction cross sections for the $3s$ state (Q_t) compared with the collisional ionization cross section (σ_i) computed for the $n = 3$ level and the total electron-capture cross sections (σ_c) from Ref. 5. Cross sections are in units of 10^{-16} cm^2 and are listed to the *nearest* whole number. (The calibration and reproducibility uncertainties in σ_i and σ_c are not shown.)

	N_2			Ar			H_2		
	10 keV	25 keV	35 keV	10 keV	25 keV	35 keV	10 keV	25 keV	35 keV
σ_i	10	10	11	12	19	16	11	7	6
σ_c	11	7	5	10	7	5	8	5	3
$\sigma_i + \sigma_c$	21	17	16	22	26	22	19	13	9
Q_t	14 ± 10	13 ± 10	12 ± 6	31 ± 10	28 ± 6	25 ± 5	19 ± 4	16 ± 3	10 ± 2

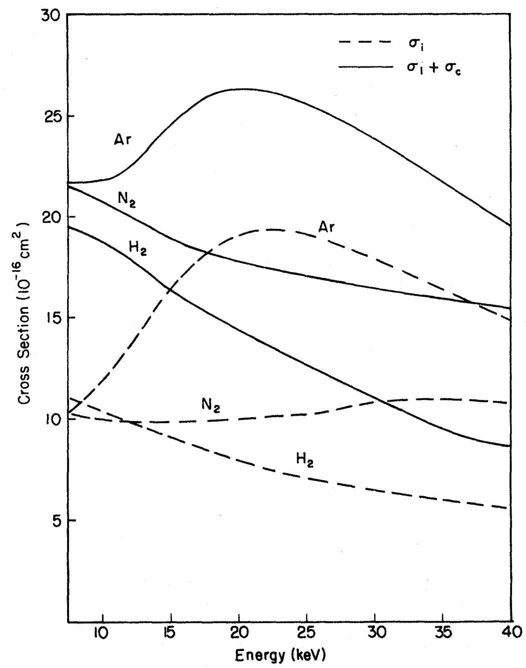

FIG. 2. Plots of the computed $n=3$ collisional ionization cross sections and the sum of these ionization cross sections and the cross sections for electron capture. This sum should be a good approximation for the destruction of all excited states.

what smaller uncertainties in H_2 are brought about by better reproducibility in the experimental data. The data for H_2 was taken at pressures about twice that used for Ar and N_2. (The H_2 pressure went to about 2 m Torr.) The reproducibility in N_2 was the poorest.

The experimental results appear to be systematically smaller than the sum of ionization and electron capture for impact on N_2 but on the other hand the results for Ar appear to be systematically larger. However, there is agreement within the estimated experimental error of the present work. It must be remembered that the computed ionization cross sections are based on electron scattering measurements performed over 40 years ago which are also subject to calibration uncertainties. Brode[2] pointed out that difference between his average curves and the measurements of the individual observers was seldom larger than 10%. (There is also an uncertainty in the σ_c values, reported to be about 5%.[8] However, the disagreement with other investigators often exceeded this.[5]) The agreement with the sum of ionization and electron capture is excellent for impact on H_2.

There is other experimental support for the concept of electron capture as an important mechanism. Donnally and Sawyer[9] found that H(2*s*) impact on Ar at 3 keV and below produced a remarkable amount of H^-. Electron impact on Ar presents a classic example of the Ramsauer effect. Although the presence of the proton may perturb the scattering potential, the Ramsauer effect should still be present from the point of view of the atomic electron. At low velocities the collisional ionization should be minimal, thus the electron-capture reaction producing H^- should dominate over the competing process of collisional ionization. It would not be surprising if large production of H^- is characteristic of low-velocity H(2*s*) impact on any gas that exhibits the Ramsauer effect.

Further evidence comes from the work of Donnally *et al.*[10] They show that electron capture may be important even when electrons are freed in the collision. They conclude from electron spin polarization measurements of electrons ejected from low-energy (0.5–1.0 keV) D(2*s*) atom impact on gases that the dominant mechanism is electron capture followed by autoionization rather than direct knockout of the electrons from the projectile atom.

A previous attempt to measure $n=3$ destruction cross sections was made by Edwards and Thomas[11] at a much higher energy. Their values are questionable, however, since they neglected collisional excitation (Q_x). This is probably a serious error at their energies of 75 to 400 keV. It is interesting to note that collisional ionization [reaction (2)] is the dominant destruction mechanism at these energies. For example, electron capture [reaction (3)] is about 10 and 2% of collisional ionization for 100 and 200-keV impact on N_2, respectively.

ACKNOWLEDGMENT

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