

Balmer- α and Balmer- β emission cross sections for $H^+ + Ar$ collisions

B. Van Zyl, H. L. Rothwell, Jr., and H. Neumann

Department of Physics, University of Denver, Denver, Colorado 80208

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Absolute cross sections for the emission of Balmer- α and Balmer- β radiations from $H^+ + Ar$ collisions between about 50- and 2500-eV proton energy and the polarizations of the emitted radiations are reported. For proton energies above 300 eV, the contributions to these radiations from decay of the long-lived 3s and 4s excited states of hydrogen are resolved. The Balmer- α emission cross section exhibits a maximum at about 1-keV proton energy. No such dominant feature is present at the $n = 4$ level of excitation. The experimental techniques used to make the measurements are described, and the results are compared with the work of other investigators and discussed on the basis of the interaction between these colliding species.

I. INTRODUCTION

In the preceding paper¹ (henceforth referred to as I), the cross sections and polarizations for the emission of Balmer-alpha and Balmer-beta radiations from $H + Ar$ collisions were reported. In this paper, we report the results of similar studies using proton projectiles. The motivations for this research are discussed in I, where the experimental techniques employed to make the measurements and the problems encountered are described in detail.

The Balmer-alpha (H_α) and Balmer-beta (H_β) radiations observed in this experiment result from the decay of excited hydrogen atoms produced by the charge exchange process



Because of the relative ease with which a proton beam may be produced (in contrast to the neutral H-atom beam used in I), numerous earlier measurements of these emission cross sections have been made. However, the present study is the first to determine the contributions to the observed radiations made by decay of the various excited states involved and to measure the polarizations of the emitted light in the low-energy region of interest here. In addition, no other H_α emission data are available for proton energies below 300 eV, and no reliable total H_β emission cross sections are available at any proton energy.

Section II discusses how the proton beam was produced and handled in this experiment and takes note of a particular problem encountered in the measurements. The results of the absolute cross section and radiation polarization measurements are presented in Sec. III, where they are compared with other data available for emissions from the $n = 2, 3$, and 4 levels of hydrogen and are discussed in terms of the basic collisional interaction.

II. EXPERIMENTAL PROCEDURE

The protons used for these studies were generated in a duoplasmatron ion source. Following extraction from the source and mass analysis, they were focused into a parallel or slightly convergent beam about 1 mm in diameter² at their point of entrance into the target cell (typically held between about 1 and 4×10^{-4} Torr Ar pressure). The target-cell arrangement used is shown in Fig. 1 of I.

Just prior to its entrance into the target cell, the proton beam was electrostatically chopped to a 50% duty cycle by deflection into a guarded ion collector. This collector, well shielded against both secondary electron and ion escape, was used to monitor the proton current during the experiment.³ Thus the proton beam entering the target cell was also square-wave modulated, allowing the same convenient separation of photon-counting signal from background as was available for the H-atom impact measurements described in I. The proton beam current was typically kept at about 0.2 μA and could be determined to within less than $\pm 3\%$ uncertainty.

In all other aspects, the $H^+ + Ar$ experiments described here were performed in the same way as the $H + Ar$ experiments described in I. The same procedures were used to measure the Ar target density and to calibrate the photon detector absolutely. Data were again taken as a function of distance into the target cell to allow separation of the contributions to the observed radiations from the various emitting hydrogen-atom states, and the polarization effects were handled in the same way.

One particular problem, however, was found to be far more severe for the case of proton impact. That is, the measured photon counting rates, when normalized to the target cell pressure, still exhibited strong increasing dependen-

cies on the target-cell pressure, indicating that second-collision effects were making substantial contributions to the measured signals.

This problem was soon identified as being a natural consequence of other processes taking place in the target cell. Since the charge exchange cross section σ_{10} for $H^+ + Ar$ is well above 10^{-16} cm² in the energy range covered here,⁴ a significant fraction (up to 20%, depending on the proton energy, target density, and target-cell position) of the incident protons can be converted to hydrogen atoms in the cell. Furthermore, since the H_α and H_β emission cross sections for H-atom impact are much larger than those for proton impact (as can be seen by comparing the results presented in I with those presented here), a substantial portion of the measured signal can be traced to this two-step mechanism.

As an example of the dependence of the pressure-normalized proton signal on the target-cell pressure, the data in Fig. 1(a) are presented. Note that, as expected, the pressure dependence of the H_β signal is much larger for those data taken near the rear of the target cell, where the longer cell path lengths afford larger opportunity for multiple-collision processes. Note also that, while not necessarily expected, the pressure-normalized signal appears to depend linearly on the target-cell pressure, allowing a linear extrapolation of the signal to zero pressure to be made at each of the target-cell positions listed, and allowing the true signals from only $H^+ + Ar$ collisions to be extracted from the measured results.

Figure 1(b) shows the same results when plotted as a function of the target cell position x , and the results obtained when zero-pressure extrapolations of the data in Fig. 1(a) are made. Note that these zero-pressure extrapolated data can be nicely fitted to the expression shown, indicating that 33% of the H_β signal from $H^+ + Ar$ collisions comes from decay of the 4s state of hydrogen and 67% from the combined decay of the 4p and 4d states. The procedures used to make this assignment are discussed in Sec. IID of I.

In general, the magnitudes of the pressure extrapolations of the photon signals increased dramatically for proton energies below 500 eV. While the total H_α and H_β emission cross sections could still be determined to within reasonable uncertainty in this lower-energy region, our confidence in assigning values to the fractions of the radiations coming from the various excited states of hydrogen becomes limited for proton energies below about 300 eV (the lowest proton energy for which such results are presented), even though such data were obtained down to 100-eV proton energy.

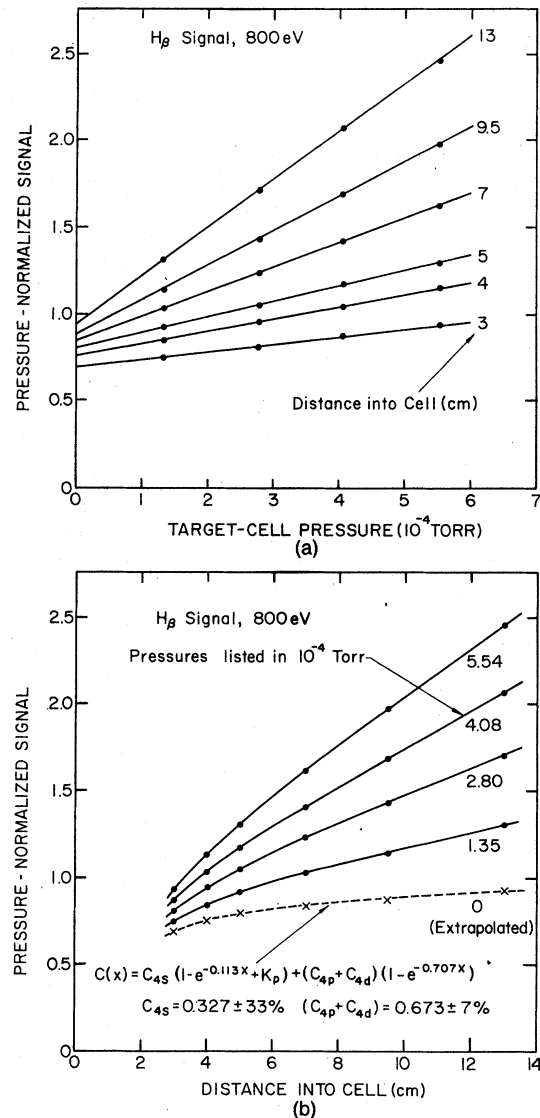


FIG. 1. (a) Target-cell pressure dependence of the pressure-normalized H_β signal at various positions in the target cell. (b) Target-cell position dependence of the pressure-normalized H_β signal at various target-cell pressures and the zero-pressure-extrapolated results.

III. RESULTS AND DISCUSSION

A. Emission-cross-section results

The H_α and H_β emission cross sections for $H^+ + Ar$ collisions are presented in Fig. 2, where they are plotted as a function of laboratory proton energy. Also shown are the fractions of the observed radiations from the long-lived 3s and 4s states of hydrogen and from the short-lived p and d states. The results have been corrected for the polarization of the emitted radiations. The total

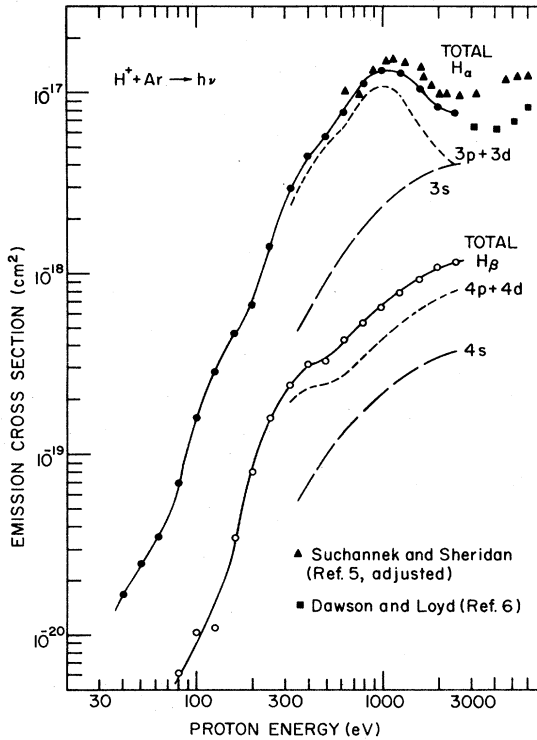


FIG. 2. H_{α} and H_{β} emission cross sections and individual state contributions.

H_{α} emission cross sections determined by Suchanek and Sheridan⁵ (adjusted as described in Ref. 5) and of Dawson and Loyd,⁶ are presented for comparison. Not shown are the data of Hess,⁷ who reports H_{α} and H_{β} emission cross sections between 0.3- and 3-keV proton energy which, while having about the same proton energy dependencies as those determined here, are about a factor of 25 smaller.

The uncertainties in the total H_{α} emission cross section average to about $\pm 16\%$ for proton energies above 200 eV and increase to about $\pm 50\%$ at 40 eV. For the total H_{β} emission cross section, the uncertainties are typically about $\pm 18\%$ down to 250 eV and increase to about $\pm 50\%$ in the 100-eV proton-energy region.

At proton energies above about 500 eV, the uncertainties in the contributions to the total radiations from decay of the short-lived p and d states of hydrogen are comparable to those cited above for the total emission cross sections. The uncertainties in the cross sections for emission from the long-lived $3s$ and $4s$ states are about twice as large, due primarily to the smaller contributions made by these states. Below 500 eV, the uncertainties in the relative contributions from the various excited states multiply rapidly as a result of the pressure-extrapolation problems

discussed above.

It is clear that the substantial maximum in the H_{α} emission cross section in the 1-keV proton-energy region comes from emission from the $3p$ or $3d$ states of hydrogen. In addition, it is highly probable that the structure in the H_{β} emission cross section at about 400-eV proton energy comes from radiation from the $4p$ or $4d$ states. Finally, even though we have insufficient confidence in the results to present them formally, the raw data contain a suggestion that radiations from the $3s$ and $4s$ states of hydrogen may become dominant in the 100-eV region, and the emergence of these contributions may explain the structures in the total emission cross sections in this low-energy range.

B. Radiation-polarization results

The polarizations of the observed H_{α} and H_{β} radiations (viewed at 90° to the proton beam axis) are shown in Fig. 3. The data presented show the net polarizations of the total emission signals. The uncertainty flags represent the quadrature combination of the measured polarization statistical uncertainties and the uncertainties associated with the data unfolding process as described in I.

Note the large negative polarization of the H_{α} radiation in the 1-keV proton-energy region, the location of the large maximum in the emission cross section shown in Fig. 2. While the H_{β} polarization is also negative in this region, it appears to swing to a positive peak at about 400-eV

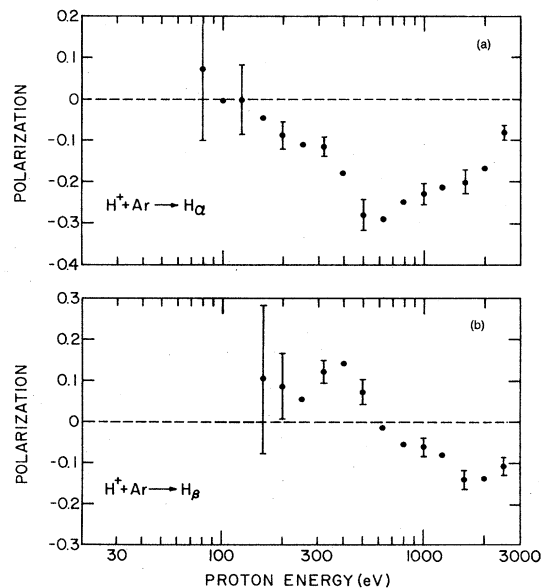


FIG. 3. Polarization of the total H_{α} and H_{β} radiations.

proton energy, the position of a definite structure in the H_β emission cross section. In fact, the H_α emission cross section also seems to show a slight structure here, which is accompanied by a rather abrupt positive trend in the H_α polarization. In the 100-eV proton-energy region, the H_α polarization seems to be near zero, supportive of the suggestion made above that radiation from the $3s$ state may be an important contributor in this region.⁸ While the uncertainty in the H_β polarization data is large at low energies, the results are not inconsistent with a similar trend.

The data presented in Figs. 2 and 3 clearly indicate that the charge-exchange-to-excited-state reactions under study here are complex, with a variety of states (and magnetic substates) being populated as a function of the incident proton energy.

C. Comparison with other excitation data

A comparison of the present results with some of the other available data for excitation of hydrogen atoms to the $n=3$ and 4 levels in $H^+ + Ar$ collisions is made in Fig. 4. The results shown are "excitation" cross sections as opposed to "emission" cross sections (that is, allowance has been made for decay branching ratios⁹ for states such as the $3p$ state which can decay via both Lyman-beta and H_α emission). Since our experiment could not separate the radiations from the $3d$ and $3p$ states of hydrogen, our lower-energy data can be described only by the cross section for excitation of the $3d$ state plus 12% of that for excitation of the $3p$ state. At the $n=4$ level of excitation, our $4s$ -state excitation cross section has been adjusted to compensate for the fractional decay of this state via Paschen-alpha emission. A similar adjustment has been made to the curve labeled $4d + (\sim 0.12)4p$, by assuming (for determining the magnitude of the adjustment required) that all the observed radiation comes from decay of the $4d$ state. Thus this curve represents the $4d$ -state excitation cross section plus only approximately 12% of the $4p$ -state excitation cross section.

The $4s$ -state excitation data shown in Fig. 4(b) clearly indicate the general proton-energy dependence of this cross section. The present results agree within uncertainty with those of Dawson and Loyd,¹⁰ who normalized their cross section to the data of Hughes *et al.*¹⁵ at about 10 keV. The results of Doughty *et al.*¹⁴ were also normalized to the work of Hughes *et al.*¹⁵ at higher energies, and the discrepancy between these sets of data in the 10–30-keV proton-energy region is a bit disturbing, since essentially the same procedures were employed for the measurements. Unfortun-

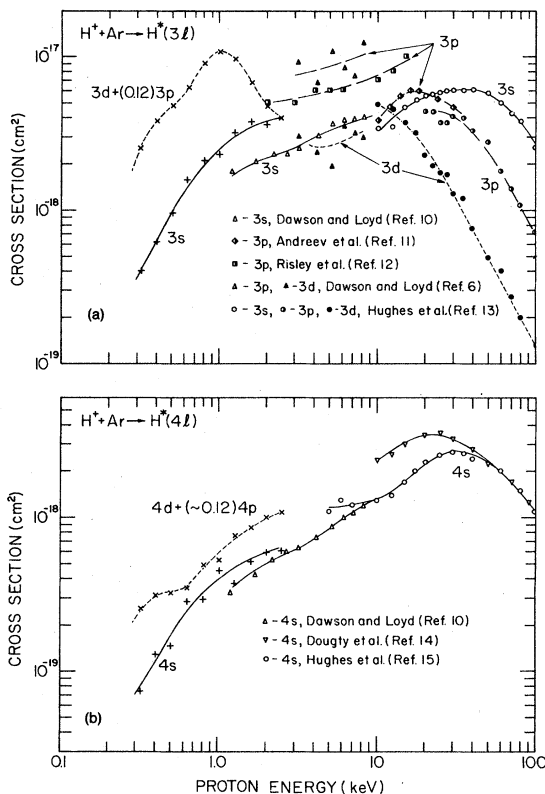


FIG. 4. Comparison of the present low-energy cross sections for excitation of the $n=3$ and 4 levels of hydrogen with other higher-energy data for $H^+ + Ar$ collisions.

ately, no other data are available for excitation to the $4p$ and $4d$ states of hydrogen.

At the $n=3$ level of excitation [Fig. 4(a)], our $3s$ -state excitation cross section is substantially above that reported by Dawson and Loyd¹⁰ and (by extrapolation) that reported by Hughes *et al.*¹³ In general, however, it appears that the $3s$ -state excitation cross section drops much less rapidly with decreasing proton energy than the $4s$ -state excitation cross section, being relatively larger in the few-keV proton-energy region.

The $3p$ -state excitation cross sections shown exhibit considerable differences in magnitude. The data of Dawson and Loyd⁶ and of Hughes *et al.*¹³ were obtained from H_α emission studies, while those of Andreev *et al.*¹¹ and Risley *et al.*¹² were obtained via observation of Lyman-beta radiation. In general, it appears that this excitation cross section reaches a maximum value near 20-keV proton energy, but does not fall rapidly with decreasing proton energy (at least down to proton energies in the 2-keV region).

On the other hand, the $3p$ -state excitation cross section appears to be smaller (or at least heading

towards a smaller value) than the $3d + (0.12)3p$ excitation cross section found here in the 1-keV proton-energy region. If it were to be assumed that the H_α emission signal measured here arises from the 12% of the decay of the $3p$ state which proceeds via H_α emission, the actual $3p$ -state excitation cross section would have to be about an order of magnitude above that reported (for example) by Risley *et al.*¹² at 2-keV proton energy. This argument leads us to conclude that the bulk of the H_α emission observed in the vicinity of the pronounced maximum in the $3d + (0.12)3p$ excitation cross section in the 1-keV proton-energy region must come from decay of the $3d$ state of hydrogen. The $3d$ -state excitation cross section thus appears to exhibit a double-maximum structure, with cross-section peaks near 1- and 10-keV proton energy.

We feel that all the cross sections reported by Hughes *et al.*¹³ (and by Dawson and Loyd,^{6,10} who normalized their results to the Hughes *et al.*¹³ data) may be too small in absolute magnitude. The data of Suchanek and Sheridan,⁵ when adjusted according to the $3s/3p/3d$ -state excitation-cross-section ratios of Hughes *et al.*¹³ and Dawson and Loyd,⁶ give total H_α emission cross sections well above the total emission cross sections reported by these workers in the 5–25-keV proton-energy region. The very-high-energy data of Ford and Thomas¹⁶ are also somewhat above the results of Hughes *et al.*¹³ A similar situation was found for the case of $H + Ar$ collisions, as discussed in I.

Taking an average of the data discussed in the previous paragraphs at the higher proton energies and using the present results at the lower energies yields the total H_α emission cross section shown by the dashed curve in Fig. 5. Shown also are some of the available Lyman-alpha (L_α) emission-cross-section data and some of the cross sections for population of the metastable $2s$ state of hydrogen in $H^+ + Ar$ collisions. Other data are available but largely duplicate those presented. The exception is the recent data of Suchanek and Sheridan²¹ for the L_α emission cross section which, while having the same proton-energy dependence as the results shown, give a cross section more than a factor of 2 smaller in absolute value.

The reason for this comparison is that the H_α emission cross section presented may be taken as a lower limit on the cascade population of the $2p$ state of hydrogen in $H^+ + Ar$ collisions. While some small fraction (~5%) of the total H_α emission goes via the $3p \rightarrow 2s$ transition, this is more than compensated by cascade population of the $2p$ state from the $n=4$ and higher levels of hydrogen in such collisions.

At the very high proton energies, the cross section for cascade population of the $2p$ state is of the same order as the "measured L_α emission cross section." This results from the fact that the cascade contribution here is mostly from higher-lying ns states, whose radiative lifetimes are sufficiently long that they escaped detection in the Hughes *et al.*¹⁹ experiment. In this proton-

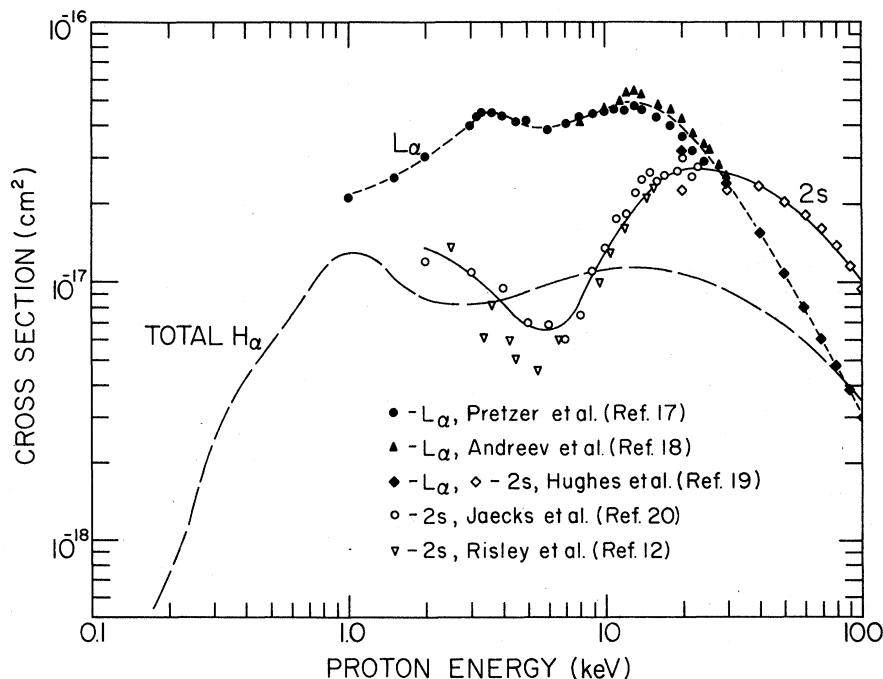


FIG. 5. L_α and H_α emission cross sections and the cross section for formation of hydrogen in the $2s$ state in $H^+ + Ar$ collisions.

energy range, the reported L_α emission cross section is thus probably reasonable close to the true $2p$ -state excitation cross section.

On the other hand, in the 1-keV proton-energy region, it appears that more than half of the observed L_α emission may result from cascade population of the $2p$ state from the $3d \rightarrow 2p$ transition, giving a true $2p$ -state direct excitation cross section below that for excitation of the $3d$ state. Furthermore, if the lower values of the L_α emission cross section reported by Suchanek and Sheridan²¹ should be more nearly correct, the effects of such cascade processes would be even more pronounced.

The effects of such cascade processes on the measured cross sections for electron capture into the metastable $2s$ state of hydrogen can be even more profound. The cross-section measurements reported by Jaecks *et al.*²⁰ and Risley *et al.*¹² were obtained by Stark-electric-field quenching of the $2s$ -state atoms formed in their target cells (i.e., where the applied electric field mixes the $2s$ and $2p$ states), and noting the increase in L_α emission when the field is applied. However, the same electric field will also mix the $3s$, $3p$; $4s$, $4p$; $3d$, $3p$; etc. states, opening the opportunity for decay of these higher-lying states via higher Lyman-line emissions during the "field-on" measurements. The cascade contribution to population of the $2p$ state will thus be reduced during the field-on measurement, causing the $2s$ -state excitation cross section to be significantly underestimated. It thus appears that the cross section for electron capture into the $2s$ state of hydrogen may be comparable to or even above that for capture into the $2p$ state in the 1-keV proton-energy region.

D. Discussion of the $H^+ + Ar$ interaction

While many more experimental data are available describing the production of excited hydrogen atoms in $H^+ + Ar$ collisions than are available for $H + Ar$ collisions as discussed in I, the state of understanding of such interactions is probably less advanced. Indeed, we are unaware of any definitive potential-energy curves upon which any such detailed understanding can be based.

As in the case for $H + Ar$ collisions, the reactions leading to excited hydrogen atoms in $H^+ + Ar$ collisions are all exothermic by more than 10 eV. Classically, one would thus expect that the reaction cross sections would reach maxima for proton energies above 10 keV. While many of the cross sections shown in Figs. 4 and 5 reach maxima in the 10–50-keV proton-energy region, many also exhibit secondary maxima or at least cross-section

structure at lower proton energies. The cross section for production of hydrogen atoms in the $3d$ state is largest in the 1-keV proton-energy range. Whatever specific reaction channel is responsible for this excitation, however, is apparently substantially less operative for producing $4d$ -state excited hydrogen atoms.

Charge capture into the ns states of hydrogen appears to exhibit a definite trend with the principal quantum number n in the lower proton-energy region. The cross section for $4s$ -state formation drops rapidly with proton energy from its maximum in the 30-keV region with only a slight structure in the vicinity of 1-keV proton energy. The $3s$ -state formation cross section, however, exhibits a more pronounced shoulder in the 1-keV proton-energy region, while that for $2s$ -state production appears to reach a definite secondary maximum somewhere below 2-keV proton energy. It appears unlikely that the small differences in exothermicity involved in these reactions can, by themselves, explain this behavior.

Risley *et al.*¹² have suggested that reactions leading to excited hydrogen atoms in $H^+ + Ar$ collisions can be interpreted on the basis of a single-electron molecular promotion model. In their picture of the collision (in which a $3p^5$ core of electrons in Ar is frozen), the first step is a charge exchange between the initial $H^+ + Ar(3p^6)$ state ($^1\Sigma$) and the $H(1s) + Ar^+(3p^5)$ state (also $^1\Sigma$). As the collision proceeds, the latter state approaches a $K^+(3p^5 3d)$ configuration of the united atom along the $^1\Sigma$ curve. At some small internuclear separation, a rotational transition ($^1\Sigma \rightarrow ^1\Pi$) occurs causing the system to separate along the $H(2p) + Ar^+(3p^5)$ state $^1\Pi$ curve. Additional rotational transitions and/or long-range (Demkov) interactions are then invoked to explain population of the $2s$ and higher-lying states of hydrogen on the outward-bound leg of the collision.

Such a model appears to have several attractive features. First, if the initial charge exchange step more or less limits the total reaction process at the lower proton energies, one might expect that the proton-energy dependencies of our measured emission cross sections would reflect that of the total charge exchange cross section.⁴ Indeed, this is approximately true for proton energies below about 1 keV, where both cross sections fall rapidly with decreasing proton energy.

Second, if the interacting system does approach the $K^+(3p^5 3d)$ united-atom configuration along the $^1\Sigma$ curve, a $^1\Sigma \rightarrow ^1\Delta$ rotational transition could also occur at small separation, causing the system to separate into the $H(3d) + Ar^+(3p^5)$ state at infinity.²² Thus it might not be unreasonable to expect that the cross sections for formation of

excited hydrogen atoms in the $2p$ and $3d$ states would be about comparable in magnitude at the lower proton energies. As can be seen in Fig. 5, in the 1-keV proton-energy region, the H_α emission cross section (mostly from decay of the $3d$ state) is about half as large as the L_α emission cross section (basically the sum of the cross sections for capture into the $2p$ and $3d$ states of hydrogen, the $3d$ state feeding the $2p$ state via the cascade process). Thus, the data are not inconsistent with such an expectation.

Furthermore, as noted in I, the polarizations of the radiations from decay of the $m_l = 0, \pm 1$, and ± 2 magnetic substates of the $3d$ state of hydrogen should be $+0.48$, $+0.26$, and -0.70 , respectively. Since our measured H_α polarization value in the 1-keV proton-energy region is about -0.3 , it would appear that a substantial part of the total H_α radiation from decay of the $3d$ state must come from the $m_l = \pm 2$ magnetic substate. This fact is consistent with the suggestion that the outgoing molecular channel during the collision is $^1\Delta$ in character.

On the other hand, Risley *et al.*¹² argue that their model successfully explains their finding that the cross section for capture into the $2p$ state of hydrogen is much larger than that for capture into the $2s$ state at the lower proton energies (these workers investigated a region down to 2-keV proton energy). As discussed in Sec. IIC, however, when the effects of cascade processes on the measurements are accounted for, it appears that the $2s$ -state capture cross section will be comparable to that for $2p$ -state capture in the 1-keV proton-energy region. From this viewpoint, the model (or at least their interpretation of the model) appears to be inconsistent with observations.

In addition, we question the use of any single-electron model in analysis of collisions of this type. Indeed, it seems possible that at least one molecular state involving the promotion of two electrons may have some influence on the various reaction channels which lead to excited hydrogen atom (and argon atom) formation.

In I, we postulated that the transient existence of a basically Coulomb state ($H^- + Ar^+$) during an $H + Ar$ collision could be used to explain a number of cross section data. To our surprise, very crude estimates of the potential-energy curves for the $H^+ + Ar$ system suggest that a similar Coulomb state ($H^+ + Ar^{2+}$) may have some influence on the reactions under discussion here.

Figure 6 shows the results of our crude estimates. For internuclear separations beyond about 2 Å, the curves shown should be fairly accurate but, with the exception of the lowest two

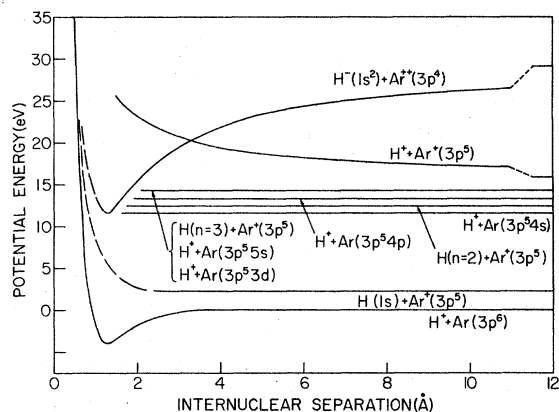


FIG. 6. Estimated potential-energy curves for the ArH^+ molecule.

curves,²³ no meaningful calculations of the potential energies of the states shown are available. The $H^- + Ar^{2+}$ curve is our estimate, drawn here to have a similar well shape but twice the depth of that presented for the $H^- + Ar^+$ state shown in Fig. 8(a) of I.

The important thing to note is that the deep minimum in the $H^- + Ar^{2+}$ potential-energy curve falls at about the same potential energy as the curves leading to excited hydrogen and argon atoms in the 1.5-Å separation region. Thus, similar to the situation described in I, the molecular wavefunctions for these excited states will probably take on ionic characteristics in the region inside about 2-Å separation. It is therefore apparent that any serious calculations of these molecular potential-energy curves will have to include the Coulomb state in the basis set employed.

Unfortunately, and in contrast to the $H + Ar$ collisions discussed in I, insufficient (low-energy) cross-section data are available to support or disprove the supposition that a transient Coulomb state may have an influence on the reactions which lead to excited atoms (or Ar^+) in $H^+ + Ar$ collisions. Interestingly, the $H^- + Ar^{2+}$ production cross section for proton energies above 2 keV does appear to exhibit structure²⁴ (perhaps analogous to that for $H^- + Ar^+$ production for H-atom impact). Furthermore, the cross section for Ar ionization, i.e., $H^+ + Ar \rightarrow H^+ + Ar^+ + e^-$ (perhaps similar to the $H + Ar \rightarrow H^+ + Ar + e^-$ reaction), does not appear to be falling rapidly with decreasing proton energy in the 5–10-keV region.²⁵ Thus it is possible that the reaction sequence $H^+ + Ar \rightarrow H^- + Ar^{2+} \rightarrow H^+ + Ar^+ + e^-$, with the first transition coming at some small internuclear separation (perhaps through a $H + Ar^+$ intermediary step) and the second at the crossing between these latter states at about 3.3

\AA , may be occurring. A critical test of this model would be to obtain additional $\text{H}^- + \text{Ar}^{2+}$ and $\text{H}^+ + \text{Ar}^+ + e^-$ cross-section data at the lower proton energies.

It would also be interesting to determine whether the $\text{H}^- + \text{Ar}^{2+}$ molecular state is populated directly via transition from the initial $\text{H}^+ + \text{Ar}$ state, or via a (charge exchange) $\text{H} + \text{Ar}^+$ intermediary state. Such a determination could be facilitated by study of $\text{Ar}^+ + \text{H}$ collisions at the lower energies.

In summary, reactions leading to excited hydrogen and argon atoms and Ar^+ ions in $\text{H}^+ + \text{Ar}$ collisions appear to be rather complex in character and probably proceed through one or more tran-

sient molecular states existing during the interaction. Insufficient data are presently available to detail the mechanism(s) involved in such interactions.

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