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Lyman-α emission from low-energy H impact on rare-gas atoms

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Absolute cross sections for the emission of Lyman-α radiation have been measured for low-energy H impact on rare-gas-atom targets. Data were obtained for H energies from 2.5 keV down to about 0.05 keV, and include measurements of the Lyman-α polarization. Using previously measured cross sections for Balmer-α and Balmer-β emission for the same collisions, it was possible to approximately evaluate the cascade contributions to the measured Lyman-α signals, and to extract the cross sections for direct $2p$-state excitation during the collisions. The results are compared with the work of other investigators where possible.

I. INTRODUCTION

In a recent paper,[1] excitation of low-energy hydrogen atoms (H) in collisions with Ne targets was described in considerable detail. Absolute cross sections for emission of the Lyman-α (L_α), Balmer-α (H_α), and Balmer-β (H_β) radiations, and the radiation polarizations, were reported for H energies from 2.5 kev down to about 0.05 keV. The results of model calculations were also described, showing how these data could be used to separate the total L_α-emission cross section into the cross sections for direct excitation to, and cascade population of, the $2p$ state of hydrogen.

In the present paper, the results of similar measurements of the L_α-emission cross sections for H impact on the other rare-gas-atom targets are reported. Also presented are the results of similar model calculations indicating how the total observed L_α emission can be divided into its direct $2p$-state excitation and cascade $2p$-state population sources. These calculations are based upon the H_α- and H_β-emission cross sections for the reactions, which have now all been measured.[2–4]

The techniques used to make the measurements reported here, including the all-important procedure for absolute calibration of the L_α photon detector, are identical to those used for the H + Ne investigations,[1] and will not be reviewed in this paper. Similarly, the model calculations, the assumptions on which they are based, and their dependence on various scaling-law parameters will not be reviewed. However, the results of some of the H + Ne studies will be presented again here, to allow a smooth following of the cross-section data through the complete sequence of rare-gas-atom targets.

The emission of L_α radiation resulting from H impact on rare-gas atoms has been the subject of several other investigations. In general, however, only the measurements of Birely and McNeal[5] extend down into the range of H impact energies of interest here, and even these data terminate at 1.0-keV H energy. Indeed, with the exception of He targets, where the recent measurements of Grosser and Krüger[6] extend down to an H energy of 0.02 keV, little experimental information about the L_α-production mechanisms operative in such interactions is available. As will be seen, again with the exception of He targets, the role played by cascade transitions to the $2p$ state from higher-lying ns and nd states populated during the interactions is much larger than would have been casually assumed, complicating the analysis of the occurring excitation processes.

II. MEASUREMENT RESULTS AND DISCUSSION

In the figures below, the results of the present L_α-emission studies are shown by the circular data points. These values are the "apparent" L_α-emission cross sections measured by viewing the L_α from the collisions at a distance of 4.3 cm into the target cell,[1] and are henceforth denoted by $Q_m(L_\alpha)$. However, because of the long radiative lifetimes of the excited ns states of hydrogen ($>1.5\times10^{-7}$ sec), some of the L_α produced by the $ns\rightarrow2p\rightarrow1s$ cascade decay sequence from the rapidly moving excited H atoms will escape detection at only 4.3 cm into the target cell. (The same is true for the decay sequence $nd\rightarrow2p\rightarrow1s$, for those transitions originating from the higher nd levels.) Thus, these $Q_m(L_\alpha)$ represent only lower limits on the true emission cross section $Q_{\rm em}(L_\alpha)$, shown by the upper solid-line curves in the figures.

Also shown in these figures by solid-line curves are $Q_{\rm cas}(2p)$, the effective cross sections for cascade populations of the $2p$ state from higher-lying ns and nd states occupied during the interactions. The contributions to $Q_{\rm cas}(2p)$ from $ns\rightarrow2p$ and $nd\rightarrow2p$ transitions are shown separately as the dashed-line curves labeled by these transitions. (These data represent the products of the nl-state excitation cross sections, deduced from the H_α- and H_β-emission cross sections,[1–4] and their appropriate $nl\rightarrow2p$ decay branching ratios, summed over $3\leq n\leq10$.) Together with the radiative lifetimes of each excited nl state, these data were used to determine the fractions of the L_α emission from cascade processes which escaped observation during the $Q_m(L_\alpha)$ measurements so that $Q_{\rm em}(L_\alpha)$ could be suitably determined. Finally, the cross sections $Q_{\rm ex}(2p)$ for direct excitation to the $2p$ state during the interactions were determined from $Q_{\rm em}(L_\alpha)-Q_{\rm cas}(2p)$, and are plotted in each of the figures as solid-line curves.

With the exception of a few scattered data points (usually at very low H energies), the uncertainties in $Q_m(L_\alpha)$ are between $\pm 14\%$ and $\pm 15\%$, largely reflecting the uncertainty in absolute calibration of the L_α-photon detector.[1] The uncertainties in $Q_{cas}(2p)$ result primarily from three sources. First, there are the uncertainties (typically about $\pm 15\%$) in the cross-section measurements for the H_α and H_β emissions for the reactions.[1-4] Second, there are the uncertainties in separating these total emissions into their $ns \rightarrow 2p$, $np \rightarrow 2s$, and $nd \rightarrow 2p$ emission components. (The measured data provide only separation of the $ns \rightarrow 2p$ component from the sum of the other two components.) This is necessary because the $np \rightarrow 2s$ component does not lead to cascade population of the $2p$ state. This component, however, is generally estimated[1] to be quite small (usually less than 10% of the total emission), because of the very small branching ratios (about 0.12) for $np \rightarrow 2s$ decay. Finally, there are the uncertainties in the model calculations[1] made to extrapolate the cross sections for excitation to the $n \geq 5$ levels from the $n = 3$ and 4 levels. Even though these uncertainties are taken to be quite large ($\pm 40\%$ to $\pm 50\%$ for $n = 5$, and $\pm 100\%$ for $n \geq 6$), the contribution made to the total $Q_{cas}(L_\alpha)$ from these higher n levels is typically $\leq 10\%$.

The uncertainties in the computed $Q_{em}(L_\alpha)$ are typically found to be very close to those assigned to the measured $Q_m(L_\alpha)$. However, the uncertainties in $Q_{ex}(2p) = Q_{em}(L_\alpha) - Q_{cas}(2p)$ vary considerably from one reaction to another. (The bulk of the uncertainties in these L_α-emission and Balmer-line-emission quantities were combined in quadrature, being judged to be uncorrelated.) When $Q_{cas}(2p)$ is very small compared to $Q_{em}(L_\alpha)$ as is found for H + He collisions, the uncertainties in $Q_{ex}(2p)$ are close to those found for $Q_{em}(L_\alpha)$. However, when $Q_{cas}(2p)$ is a large fraction of $Q_{em}(L_\alpha)$ as in H + Ar collisions, the uncertainties in $Q_{ex}(2p)$ become quite large as well. It would be impractical here to specify the uncertainty in each of these computed cross-section curves at each H energy, but typical values will be cited as the data are presented.

The experimental results for $Q_m(L_\alpha)$ obtained by Birely and McNeal[5] and by Orbeli *et al.*[7] for H impact energies below 10 keV are also presented. These investigators also appear to have made measurements with detectors situated to view L_α from interactions occurring some 4 or 5 cm into their respective target cells. Thus, comparisons of their $Q_m(L_\alpha)$ with those reported here should be reasonably valid, at least to within the extent that the respective absolute calibrations of the L_α detectors employed are similar.

Orbeli *et al.*[7] calibrated their detector by observing L_α from H^+ + Ne collisions, using the measured L_α-emission cross section of Andreev *et al.*[8] for this reaction as a standard. In contrast, Birely and McNeal[5] used the L_α-emission cross section for H^+ + Ar collisions reported by Pretzer *et al.*[9] as their standard, although they note the close agreement between this result and the H^+ + Ar data of Andreev *et al.*[8] Indeed, these two measurements are well within mutual uncertainties for H^+ energies in the 10–25 keV range (where the Pretzer *et al.*[9] data average to about 6% below the Andreev *et al.*[8] results). In addition, the data of Pretzer *et al.*[9] merge smoothly onto the L_α-emission cross section for H^+ + Ar collisions measured in this laboratory[1] for H^+ energies below 3 keV.

While there is, therefore, no reason to believe that all these L_α-detector calibrations are substantially different, things are not as nice as they could be. Unfortunately, the Pretzer *et al.*[9] data were never corrected to account for an increasing loss of L_α signal with increasing H^+ energy, due to the effect of Doppler shifting of the L_α wavelength on the transmission of the narrow-bandpass O_2 gas filter used to isolate the L_α signal during the measurements.[10] Application of such a correction necessitates increasing the Pretzer *et al.*[9] cross-section values by about 6% for H^+ energies near 10 keV and close to 20% for H^+ energies near 25 keV.

In addition, the data of Pretzer *et al.*[9] were taken by viewing L_α from the collisions at a distance of only about 1.7 cm into their target cell. Because Birely and McNeal[5] viewed the L_α at close to 5 cm into their target cell, a significantly larger fraction of the L_α from the $ns \rightarrow 2p \rightarrow 1s$ cascade decay sequence was observed. The $Q_m(L_\alpha)$ values of Birely and McNeal[5] should thus have been normalized to slightly larger values (by perhaps 5–10%) than those determined by Pretzer *et al.*[9] The combination of these two effects suggest that the $Q_m(L_\alpha)$ data of Birely and McNeal[5] shown here should be revised upward by approximately 20%.

A. H + He collisions

The absolute cross sections for L_α emission resulting from H impact on He targets are shown in Fig. 1. As noted above, the present $Q_m(L_\alpha)$ are typically uncertain by between $\pm 14\%$ and $\pm 15\%$ for H energies above 0.05 keV (becoming about twice as uncertain at 0.032 keV). The estimated uncertainties in $Q_{cas}(2p)$ average here to be about $\pm 25\%$ (between $\pm 15\%$ and $\pm 20\%$ for the $ns \rightarrow 2p$ component, and $\pm 30\%$ to $\pm 35\%$ for the $nd \rightarrow 2p$ component). However, because $Q_{cas}(2p)$ is here so small, the uncertainties in $Q_{em}(L_\alpha)$ and $Q_{ex}(2p)$ are also small, averaging to only about $\pm 14\%$ and $\pm 16\%$, respectively.

As seen in Fig. 1, the present $Q_m(L_\alpha)$ lie about 20% above the values reported by Birely and McNeal,[5] possibly reflecting the absolute L_α-detector calibration problem discussed above. However, increasing the $Q_m(L_\alpha)$ of Birely and McNeal[5] by 20% causes their values to be even further above those of Orbeli *et al.*[7] and of Dose *et al.*[11] (The difference between these latter two measurements is somewhat surprising, because both L_α detectors were calibrated against the data of Andreev *et al.*[8] for H^+ impact on rare-gas-atom targets.) Consideration of such higher-H-energy data as reported by Hughes and Choe,[12] for example, sheds little light on this dilemma, for their results, while in good agreement with those of Birely and McNeal,[5] were also normalized absolutely to the data of Pretzer *et al.*[9] for L_α emission from H^+ impact on rare-gas atoms.

Also shown in Fig. 1 are the data of Grosser and Krüger[6] for H energies below about 1 keV. These workers examined the dependence of the L_α from the collisions as

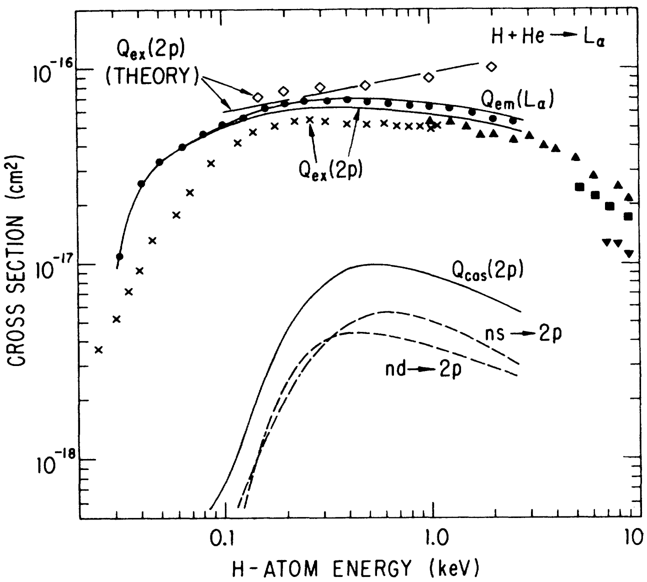

FIG. 1. L_α-emission cross sections for H + He collisions. The measured data shown are from the following: ●, present results; ▲, Birely and McNeal (Ref. 5); ■, Orbeli *et al.* (Ref. 7); ▼, Dose *et al.* (Ref. 11); and ×, Grosser and Krüger (Ref. 6). The theoretical results are from: ◇, Benoit and Gauyacq (Ref. 22); and the long-dashed curve, Bell *et al.* (Ref. 21).

a function of an electric field applied in their target cell and, via an analysis, were able to obtain the plotted $Q_{ex}(2p)$ values. It can be seen that their data, at least for H energies above 0.1 keV, exhibit an H-energy dependence essentially identical to that obtained here. However, once again the absolute L_α-detector calibration problem becomes significant.

Grosser and Krüger[6] normalized their L_α-emission data for H + He collisions to those of Birely and McNeal,[5] who as noted above, calibrated their results to the H^+ + Ar results of Pretzer *et al.*[9] The uncertainties in the data of Pretzer *et al.*[9] were already cited as ±45%, reflecting a tortuous path of L_α-emission intercomparisons involving five calibration transfers between various reactions back through the work of Dunn *et al.*[13] and Fite and Brackmann,[14] whose measurements of L_α emission from electron-H atom collisions were, in turn, normalized to theoretical predictions.[15–17] We thus believe that there is adequate justification to suggest that these results of Grosser and Krüger[6] (and, for that matter, those of Birely and McNeal[5]) be adjusted upward by about 20% based upon the present $Q_m(L_\alpha)$ data.

It is not the purpose of this paper to discuss in detail the theoretical calculations available for H-atom excitation during the interactions of interest here. Indeed, such older calculations as made, for example, by Levy,[18] Flannery,[19] or Bell *et al.*[20] for H + He collisions are completely inadequate in describing the experimental results at lower H energies, due to neglect of the molecular nature of the interactions. (Some comparisons of these calculations with experimental data have been given by Birely and McNeal[5] and by Bell *et al.*[21])

While we question the merit of looking at all such interactions from the viewpoint of one-electron promotion, molecular-correlation diagrams, the H + He→L_α reaction seems an ideal candidate for such an analysis. The strong binding of the $1s^2$ electrons to the He atom probably justifies the assumption that they remain in their core during the interaction. Furthermore, the data shown in Fig. 1 indicate that most ($\gtrsim$85%) of the L_α resulting from the interactions comes from direct excitation to the $2p$ state of hydrogen, and not from cascade processes.

Bell *et al.*[21] and Benoit and Gauyacq[22] have calculated $Q_{ex}(2p)$ for H + He collisions under the assumption that the operative excitation mechanism can be approximated by a two-state rotational-coupling model. The states involved are shown schematically in Fig. 2, where it has been assumed that the He($1s^2$) core is frozen as the incident atoms approach the united-atom limit. At an internuclear separation of about $1a_0$, according to these models, a rotational transition occurs from the incident $2p\sigma$ molecular orbital (MO) to the outgoing $2p\pi$ MO leading to population of the hydrogen $2p$ state. The possible radial (translational) coupling leading to a $2p\sigma$-$2s\sigma$ transition at the (diabatic) crossing of these curves is ignored, because this crossing presumably occurs at a much smaller internuclear separation.[22]

The results of these calculations are also shown in Fig. 1. As can be seen, at least for H energies below about 0.6 keV, these results are within about 25% of the $Q_{ex}(2p)$ obtained here. This overestimate of the cross section might be expected due to ignoring second-step rotational transitions from the $2p\pi$ MO back down to the $2s\sigma$ MO on the outward leg of the interactions. (Macek and Wang[23] have recently shown that consideration of such two-step rotational couplings is needed to explain product-H-atom excitation in H^+ + He collisions, where the same MO's shown in Fig. 2 are involved.) Such calculations may thus more nearly reflect $Q_{ex}(2p)+Q_{ex}(2s)$,

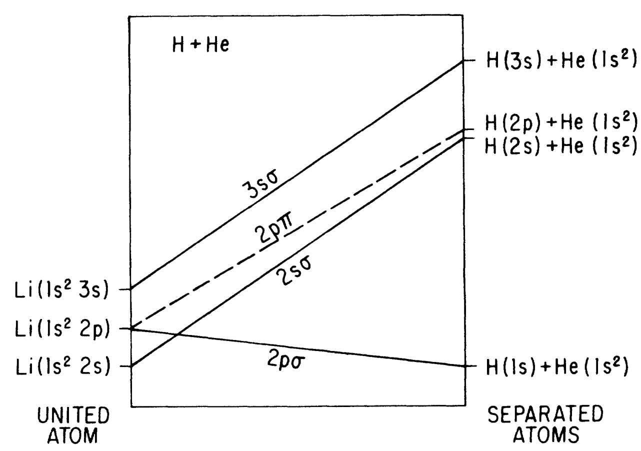

FIG. 2. Schematic diabatic correlation diagram for H + He collisions.

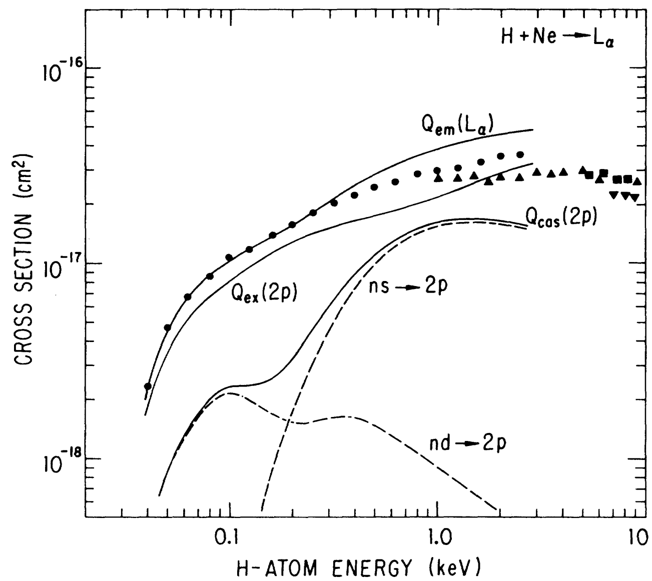

FIG. 3. L_α-emission cross sections for H + Ne collisions. The measured data shown are from the following: ●, Van Zyl *et al.* (Ref. 1); ▲, Birely and McNeal (Ref. 5); ■, Orbeli *et al.* (Ref. 7); and ▼, Dose *et al.* (Ref. 11).

consistent with the fact that for H + He collisions, the cross-section ratio $Q_{ex}(2s)/Q_{ex}(2p)$ is about 0.25 at the lower H energies.[5,24] The data presented here are, therefore, in reasonable agreement with this interpretation of the interactions.

B. H + Ne collisions

The data for L_α emission from H + Ne collisions are shown in Fig. 3. (The uncertainties in these data were discussed earlier,[1] and will not be reviewed here.) Note that the present $Q_m(L_\alpha)$ values again lie about 20% above those of Birely and McNeal.[5] Here, however, the data of Orbeli *et al.*[7] and Birely and McNeal[5] are in essential agreement, and those of Dose *et al.*[11] are less discrepant than for the case of He targets. This suggests that other measurement parameters (for example, target-density determinations) may as well be accounting for some of the discrepancies among the results, as opposed to identifying all such differences as due to the relative L_α-detector calibrations used for the measurements.

The prime reason for reviewing these data here, however, is to point out how different they are from the case of He targets shown in Fig. 1. Indeed, at the lower H energies, $Q_{ex}(2p)$ is almost an order of magnitude smaller than for He targets. The $ns \rightarrow 2p$ component of $Q_{cas}(2p)$ is almost comparable to $Q_{ex}(2p)$ for H energies in the 1–2-keV range, accounting for about 40% of the total L_α emission (as opposed to less than 10% for He targets). This enhanced relative importance of *ns*-state excitation in H + Ne collisions was also noted by Birely and McNeal.[5] Only for this reaction does their apparent $Q_{ex}(2s)$ exceed their $Q_{ex}(2p)$ at the lower H energies, in agreement with measurements made earlier in this laboratory.[1]

The H + Ne interaction can also be examined with the aid of a one-electron promotion, molecular-correlation diagram under the assumption that the $1s^2 2s^2 2p^6$ Ne electrons retain their core configuration during the collisions. For Ne, this is probably still a reasonable assertion, because the outer-shell $2p$ electrons are bound by 21.6 eV, about 8 eV more than the H atom's electron. Figure 4 shows the resulting diabatic molecular-correlation diagram,[25] indicating that the incident interaction channel takes the form of a $3d\sigma$ MO.

A rotational coupling ($3d\sigma$-$3d\pi$) is again possible here to cause hydrogen excitation to the $2p$ state. However, the incident $3d\sigma$ MO must cross the $3p\sigma$, $3s\sigma$, and $4s\sigma$ MO's leading, respectively, to H(2*s*), H(3*s*), and H(4*s*) formation, and even the $3p\pi$ MO leading to H(3*p*) formation, before approaching the Na($2p^6 3d$) united-atom limit. While such diagrams give little information about where these various crossing occur, the first few probably occur at sufficiently large internuclear separations that radial transitions to the excited *ns* states of hydrogen will be much more probable than at the $2p\sigma$-$2s\sigma$ crossing in H + He interactions shown in Fig. 2. Thus, there is at least qualitative agreement between this picture and the relatively large $Q_{ex}(ns)$ and small $Q_{ex}(2p)$ found here for the case of H + Ne interactions.

C. H + Ar collisions

Figure 5 shows the results of the studies made for the case of Ar targets. The measured $Q_m(L_\alpha)$ are again uncertain by about ±15% for H energies above 0.04 keV, and again lie about 20% above those of Birely and McNeal.[5] Here, however, the data of Orbeli *et al.*[7] and

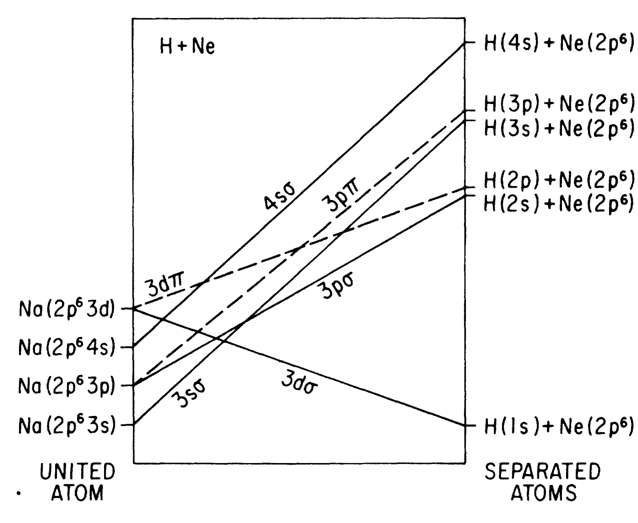

FIG. 4. Schematic diabatic correlation diagram for H + Ne collisions.

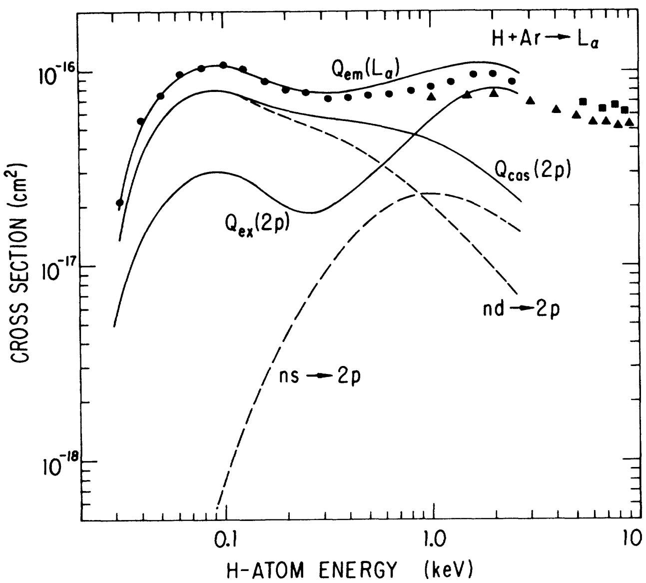

FIG. 5. L_α-emission cross sections for H + Ar collisions. The measured data shown are from the following: ●, present results, ▲, Birely and McNeal (Ref. 5); and ■, Orbeli *et al.* (Ref. 7).

the present results are in surprising agreement, although the data of Dose *et al.*[11] (not shown here) are well below the other values. The uncertainty in $Q_{em}(L_\alpha)$ is typically also again about ±15%. At the higher H energies, the uncertainties in both $Q_{cas}(2p)$ and $Q_{ex}(2p)$ lie in the ±15% to ±20% range.

The overwhelming feature of these results is clearly the very large contribution made to the total L_α emission by the $nd \rightarrow 2p$ component[2] of $Q_{cas}(2p)$ at the lower H energies. Indeed, for H energies below about 0.4 keV, Fig. 5 shows that about 70% of the total L_α emission comes from this cascade process. The effect of this large cascade contribution, however, and its estimated uncertainty of about ±20%, causes $Q_{ex}(2p)=Q_{em}(L_\alpha)-Q_{cas}(2p)$ to become increasingly uncertain with decreasing H energy, being close to ±70%, for example, at 0.1 keV H energy.

Some confirmation of the (relative) smallness of $Q_{ex}(2p)$ at the lower H energies is provided by the recent studies of H + Ar collisions made by Grosser and Krüger.[26] These workers do not claim a definitive determination of $Q_{ex}(2p)$ in the spirit of their He-target work,[6] but their measured $Q_{ex}(2p)+Q_{ex}(3s)+Q_{ex}(3d)$ is consistent with the fact that $Q_{ex}(2p)$ falls rapidly with decreasing H energy below 1 keV. In a subsequent report,[27] the value $Q_{ex}(2p)=2.9\times10^{-17}$ cm^{-2} was given for an H energy of 0.3 keV. This value is somewhat above that obtained here (2.0×10^{-17} cm^2), but well within the estimated mutual uncertainties for the two determinations.

For the cases of H impact on Ar, Kr, or Xe, the binding energies of the target's outer-shell electrons are close to that for the H atom. In our opinion, there is, therefore, little justification for attempting to describe such interactions by a model allowing promotion of only the H atoms's electron. (It is interesting to note, however, that the incident MO's in such correlation diagrams join to increasingly higher excited states of the united alkali-metal atoms, providing opportunities for numerous rotational- and radial-coupling transitions at or near various diabatic curve crossings.)

An alternate explanation to account for the efficient *nd*-state excitation found to occur in low-energy H + Ar collisions was postulated earlier.[2,28] This involved viewing the interaction as the sequence $H+Ar \rightarrow H^- + Ar^+ \rightarrow H(nd)+Ar$, where the transient-intermediate Coulomb state provides the operative "coupling" mechanism. The final *nd*-state populations thus result from the "second" transitions occurring at (diabatic) crossings on the outward leg of the interactions at large internuclear separations (as the ion-pair potential energy slowly climbs towards its separated-ion value).

Because H^- and Ar^+ have ^1S and ^2P configurations, respectively, the Coulomb state(s) can only be $^2\Sigma$ or $^2\Pi$ in character. At large receding separations, where the intermolecular and H-beam axes become synonymous, these molecular states thus reduce to the $m_l=0$ and ±1 sublevels of the excited *nd* states, but the $m_l=\pm2$ sublevels would not be populated (because they would come from a $^2\Delta$ molecular state). This would account for the large positive polarizations of the H_α and H_β emissions observed for H + Ar collisions at the low H energies,[29] as well as other features of the data.[2] That the *nd* states would be preferentially populated by this mechanism is realistic, because the $3d_0$ sublevel in particular, has a highly extended wave-function lobe along the internuclear axis, providing for its ease of population at the large internuclear separations. (As the H^- and Ar^+ recede, the loosely bound H^- electrons are probably concentrated between the H^+ and Ar^+ cores, similar perhaps to a distorted H_2 molecule.)

The $2p_0$ wave function, of course, also exhibits a lobe along the receding internuclear axis in this picture of the interaction. Thus, it would not be surprising if a similar $2p$-state-population mechanism would be operative in H + Ar collisions. The fact that $Q_{ex}(2p)$ also exhibits a maximum at low H impact energies, as seen in Fig. 5, is certainly not inconsistent with this suggestion.

D. H + Kr collisions

The L_α-emission cross sections for H impact on Kr targets are shown in Fig. 6. As can be seen, these results (and, consequently, their uncertainties) are very similar to those shown above for Ar targets. Once again, the striking feature of these data is the very large contribution made to $Q_{em}(L_\alpha)$ by the $nd \rightarrow 2p$ component of $Q_{cas}(2p)$ at low H energies. Here, however, $Q_{ex}(2p)$ seems to again emerge as the dominant source of L_α at very low H energies. (Had this been recognized as these data were being acquired, additional measurements at still lower H energies would have been made.)

In general, if the interpretation given above for H + Ar collisions is correct, a similar interpretation should be

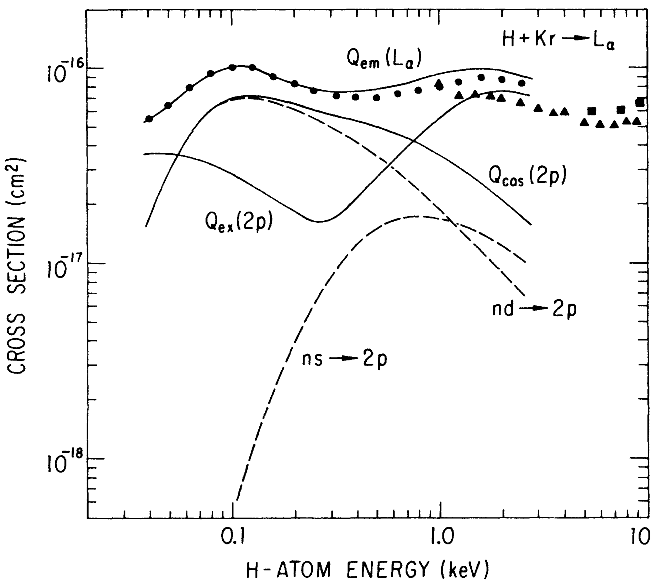

FIG. 6. L_α-emission cross sections for H + Kr collisions. The measured data shown are from the following: ●, present results; ▲, Birely and McNeal (Ref. 5); and ■, Orbeli *et al.* (Ref. 7).

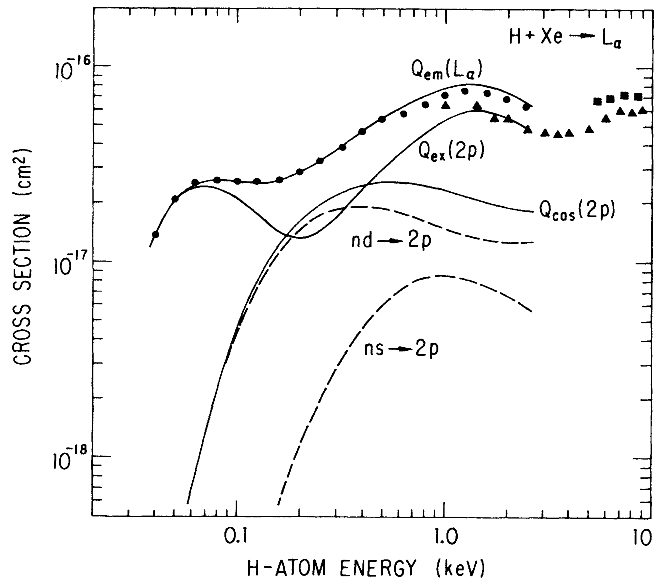

FIG. 7. L_α-emission cross sections for H + Xe collisions. The measured data shown are from the following: ●, present results; ▲, Birely and McNeal (Ref. 5); and ■, Orbeli *et al.* (Ref. 7).

applicable to these H + Kr data. The H_α and H_β emissions from this reaction are again highly positively polarized[4] at the lower H energies. It is also interesting to note that because of the lower ionization potential of Kr, the $H^- + Kr^+$ state is lower in energy than for the case of Ar targets. Thus, the $H^- + Kr^+ \rightarrow H^* + Kr$ transitions occurring on the outward leg of the interaction must take place at curve crossings at even larger internuclear separations than for Ar targets (where they already occur[2] at $\gtrsim 5a_0$).

Note once again that the present $Q_m(L_\alpha)$ values average to about 20% above those of Birely and McNeal,[5] and are again in good agreement with those of Orbeli *et al.*[7] (Dose *et al.*[11] did not examine L_α emission from H impact on Kr or Xe targets.)

For the case of H + Kr collisions (and H + Xe collisions to be discussed below), a slight possibility exists that some of the observed ultraviolet photons from the interactions were not L_α. When the O_2-gas filter used to "isolate" L_α was evacuated, the photon-counting rate increased by a factor of about 2.1 (about 2.5 for Xe targets, with both factors showing some dependence on H energy). The counting-rate increase expected for a pure L_α signal[1] was a factor of 1.79. However, while Kr and Kr^+ (and Xe and Xe^+) have emission lines[30] within the full wavelength bandpass of the unfiltered photon counter, the more intense of these emission lines (at least as produced in gas discharges[30]) do not lie close to the O_2-gas-filter transmission windows.[1] The possibility of substantial L_α-signal contamination is thus considered remote.

E. H + Xe collisions

The L_α-emission cross sections for H impact on Xe targets are shown in Fig. 7. The measured $Q_m(L_\alpha)$ are again typically uncertain by ±15%, as is the computed $Q_{em}(L_\alpha)$. Here the uncertainties in $Q_{cas}(2p)$ range between about ±20% and ±25%, as do those in $Q_{ex}(2p)$ at the higher and lower H energies, the latter being again more uncertain for H energies between about 0.125 and 0.63 keV where its magnitude is smaller than $Q_{cas}(2p)$. The $Q_m(L_\alpha)$ obtained here and by Orbeli *et al.*[7] are again in good agreement, both being again approximately 20% larger than the results of Birely and McNeal.[5]

While the $nd \rightarrow 2p$ component of $Q_{cas}(2p)$ is still a non-negligible source of L_α for this reaction at the lower H energies, it obviously makes a much smaller contribution than for the cases of Ar and Kr targets shown above. The large maxima in this contribution found for these other targets at H energies near 0.1 keV is simply not present here.[4] In contrast, $Q_{ex}(2p)$ does exhibit a maximum at the lower H energies, which is quite similar to those found for Ar and Kr targets. This finding is in support of the tentative model given above for L_α emission from H + Ar and H + Kr collisions.

Because the ionization potential of Xe is only 12.1 eV, the $H^- + Xe^+$ ground state at infinite separation lies only 11.4 eV above the initial H + Xe ground state. The Coulomb-state potential-energy curve will therefore (diabatically) cross the $H(2p)$ + Xe state (at 10.2 eV) as the ion pair separates, but will not cross the $H(3d)$ + Xe state at (12.1 eV) even at infinite separation. Thus the postulated mechanism for explaining the significant hydrogen *nd*-state excitation during low-energy H impact on Ar and Kr targets cannot occur for Xe targets, nicely consistent with the experimental data.

However, it must be noted that the above argument only applies to the ground-$Xe^+(^2P_{3/2})$ state. Because of the large doublet splitting present in this ion (about 1.3

eV), the $H^- + Xe^+(^2P_{1/2})$ Coulomb state could still cross the H(*nd*) + Xe states as the ion pair separates after its production in the reaction sequence. While this may weaken the above model to some extent, it may also suggest that the $^2P_{3/2}$ states of the Ar^+ and Kr^+ ions involved as transient species during their respective interactions are the most effective as the Coulomb-state intermediaries.

The polarizations of the measured L_α emission for H impact on all rare-gas atoms are shown in Fig. 8. These data (obtained using Brewster angle-reflection techniques[1]) were measured by observing the emitted L_α at 4.3 cm into the target cell. All the measured $Q_m(L_\alpha)$ reported above were adjusted[1] to account for these polarization data.

With the exception of a few data points for H + Xe collisions, the L_α-polarization values obtained here are all positive. To within uncertainties, the data for He targets show little variation with H energy, similar to this regard to the L_α-emission cross section itself (Fig. 1). This is not true for the Ne-target data, which exhibit the largest of all the measured polarizations found here at the lower H energies, where the L_α-emission cross section is the smallest found here (Fig. 3). The rapid drop in these polarization values with increasing H energy is consistent with the increasing contribution made to the total L_α emission by $Q_{cas}(2p)$ from $ns \rightarrow 2p$ transitions (which must result in unpolarized L_α, because of the spherical symmetry of the initial *ns*-state wave functions).

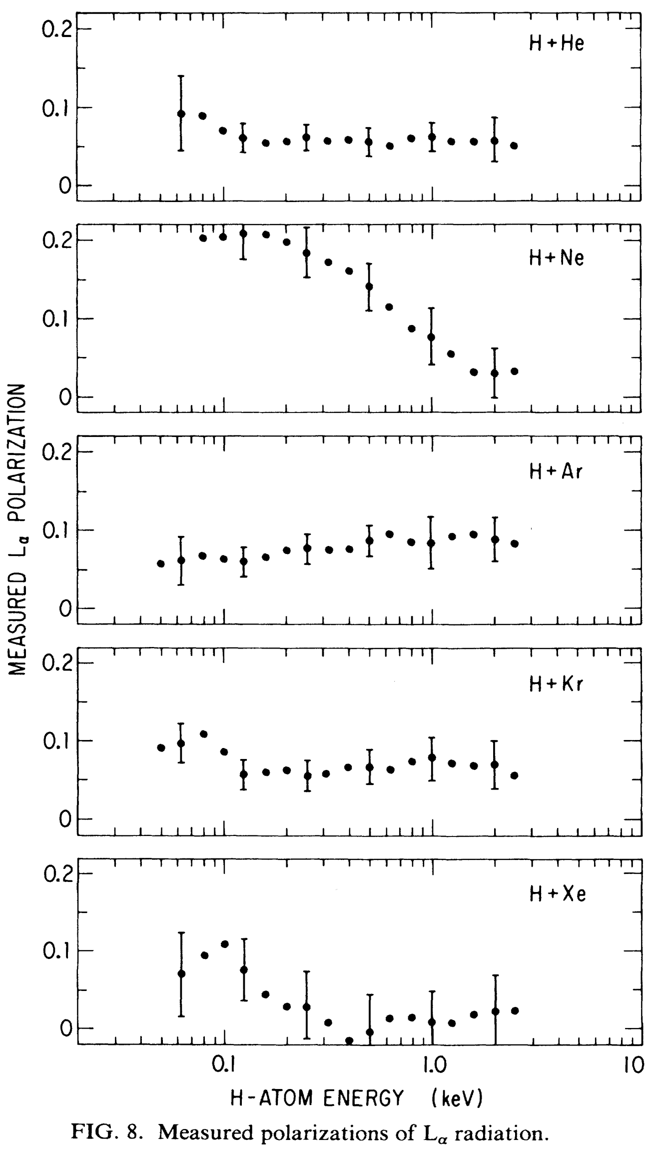

FIG. 8. Measured polarizations of L_α radiation.

The polarization data for Ar and Kr targets also show only minimal dependence on H energy. For these reactions, the L_α-emission cross sections (Figs. 5 and 6) are so heavily influenced by cascade processes that these results provide little by way of increased understanding of the interactions. However, for Xe targets, there is an indication that the L_α polarizations do increase somewhat at the lower H energies, where $Q_{ex}(2p)$ exhibits its low H-energy maximum (Fig. 7). This is certainly not inconsistent with the possibility that the $2p_0$ state is being populated here (from a Coulomb intermediate state) via the $m_l = 0$ wave-function lobe extending along the internuclear axis, as it directionally approaches the H-beam axis at increasing internuclear separation.

It should be noted that a similar interpretation of the positive L_α polarization for H + He collisions would be inappropriate. If the models of Bell *et al.*[21] and Benoit and Gauyacq[22] are correct, the collisionally receding $2p\pi$ MO would feed the $m_l = \pm 1$ sublevels of the $2p$ state, suggesting perhaps that the L_α polarization should be negative.[29] Here, however, the intermolecular axis at the time of the $2p\sigma$-$2p\pi$ transition is probably nearly perpendicular to the H-beam axis (the reference for our measured L_α polarizations). Because of the very rapid intermolecular-axis rotation occurring for such small impact-parameter interactions, the electron cloud may lag behind this rotation, giving rise to a charge distribution which is not symmetric about the internuclear axis. Furthermore, additional rotational coupling between the $2p\pi$ MO and the $3d\sigma$ MO leading to the $2p_0$ state of hydrogen from the united Li($1s^2 3d$) atom (not shown in Fig. 2) can occur on the outward leg of the interaction. Thus, the predictions of these models and the measured L_α polarization are not in direct conflict, although such models obviously oversimplify the nature of the total interaction.

In summary, it is clear that L_α emission resulting from H impact on rare-gas atoms is a complicated subject involving a variety of differing excitation channels during the interactions. While the data for He targets are basically consistent with simple theoretical predictions,[21,22] the results for the heavier atoms can only be said to be in qualitative agreement with much more poorly defined models of the interactions, inviting more detailed theoretical work. In addition, in view of the good agreement between the present measurements and those of Orbeli *et al.*[7] for L_α emission from H impact on Ar, Kr, and Xe targets, we suggest an upward revision of about 20% be made to the data of Birely and McNeal,[5] and to all other results that have used these data as a L_α-calibration standard.

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