Balmer-α and Balmer-β emission cross sections for low-energy H collisions with He and H_2

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Absolute cross sections for the emission of Balmer-α and Balmer-β radiations resulting from H + He and H + H_2 collisions are reported for hydrogen-atom energies between about 50 eV and 2.5 keV. The polarizations of the emitted radiations are also reported. For hydrogen-atom energies above 200 eV, the contributions to these radiations from decay of the relatively long-lived 3*s* and 4*s* excited states of the projectile atoms are resolved. The results of the measurements are compared with other available data for these reactions, as well as with data for H impact on other target species.

I. INTRODUCTION

The Balmer-α (H_α) and Balmer-β (H_β) emission lines of atomic hydrogen play important roles in many diverse areas of physics. Their wavelengths (656.3 and 486.1 nm, respectively) are easily observable, a fact which has enhanced their usefulness as tools to the physicist and the astronomer for observing and probing a wide range of physical phenomena.

This paper reports measurements of the absolute emission cross sections for these radiations resulting from low-energy hydrogen-atom (H) impact on He and H_2 targets, the simplest two-electron atoms and molecules, respectively. In spite of the relative simplicity of these reactions, however, no detailed theoretical understanding of the collisional excitation processes responsible for these emissions exists for H-atom energies in the keV range and below, where the collision velocity is well below the electronic Bohr velocity. It is hoped that the present results, which include resolution of the $3s \rightarrow 2p$ and $4s \rightarrow 2p$ hydrogenic-transition components of the total H_α and H_β signals, and determinations of the polarizations of the emitted radiations, will stimulate an interest in examining the collisional mechanisms operative in such excitation processes.

This work is an extension of the earlier results of Van Zyl *et al.*,[1] who reported the cross sections for production of these same emissions for the case of H + Ar collisions, and of Van Zyl and Neumann[2] for similar measurements for the cases of N_2 and O_2 targets. Because the experimental techniques used for the present studies (such as generation of the projectile H-atom beam, determination of the target-gas density, and absolute calibration of the photon detector) were essentially identical to those employed for the earlier H + Ar work, they will not be reviewed here. Just as in all these earlier investigations, those components of the total H_α and H_β signal resulting from decay of the relatively long-lived 3*s* and 4*s* excited states of the projectile H atoms were isolated from the other signal components by observation of the growth of the measured photon signal as a function of distance along the H-atom-beam axis into the target cell.

The H_α and H_β emission cross sections reported here have been corrected to account for the polarizations of the emitted radiations (observed at 90° relative to the axis of the H-atom beam). These polarizations, formally defined by $P=(I_{\parallel}-I_{\perp})/(I_{\parallel}+I_{\perp})$, where $I_{\parallel}$ and $I_{\perp}$ are the emission intensities with their planes of polarization parallel and perpendicular to the H-atom-beam axis, respectively, were determined by the same procedures used for the earlier H + Ar studies.[1]

The total H_α emission cross section for proton (H^+) impact on He was also approximately determined for H^+ energies of 2.0 and 1.25 keV. Once again, the techniques used will not be discussed here, as they were documented in detail in an earlier paper[3] describing similar measurements for H^+ + Ar collisions.

For the case of H + H_2 collisions, it is of course possible to produce hydrogen atoms excited to the *ns*, *np*, and *nd* states of interest here by dissociative excitation of the target H_2 molecule as well as by excitation of the projectile H atoms. While the photon signals from these dissociative excitation processes could not be distinguished experimentally from the signals resulting from decay of the short-lived *np* and *nd* excited states of the projectile H atoms, the signals from the longer-lived *ns* excited states of the projectile H atoms could still be isolated by the same procedures used for the other target gases.

II. RESULTS OF THE MEASUREMENTS

The results of the measurements for the case of H + He collisions are shown in Fig. 1, where the total H_α and H_β emission cross sections and the contributions to these emissions from the excited states of the projectile H atoms labeled are plotted as a function of laboratory H-atom energy. The absolute values of the total H_α and H_β emission cross sections are uncertain by about $\pm 15\%$ for H-atom energies above 80 eV, increasing to about $\pm 40\%$ for the lowest-energy data.[4] While the uncertainties associated with the contributions to these radiations from the excited states labeled are somewhat larger, only for a few of the data points showing that component of the H_β emission arising from decay of the 4*s* state, do the uncertainties exceed $\pm 20\%$.

The polarizations of the H_α and H_β radiations resulting from H + He collisions are shown in Fig. 2, where the two sets of data describe the polarizations of the total H_α and H_β emissions, and the polarizations of those parts of the emissions resulting from decay of the $np + nd$ excited

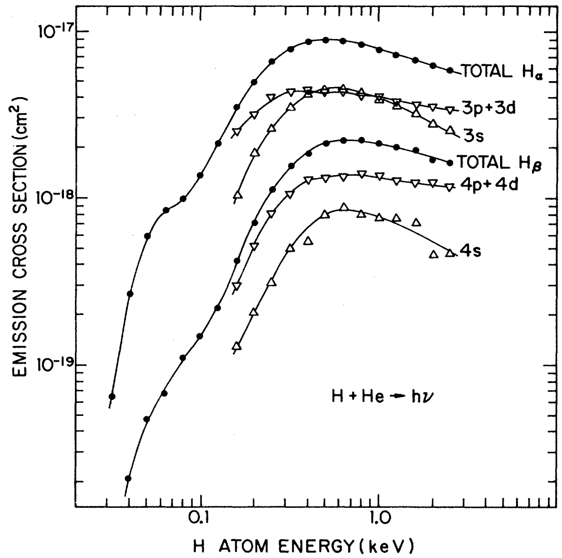

FIG. 1. Total H_α and H_β emission cross sections for the H+He reaction. Contributions to these emission cross sections from decay of the hydrogenic states listed were separated for H-atom energies above 0.16 keV.

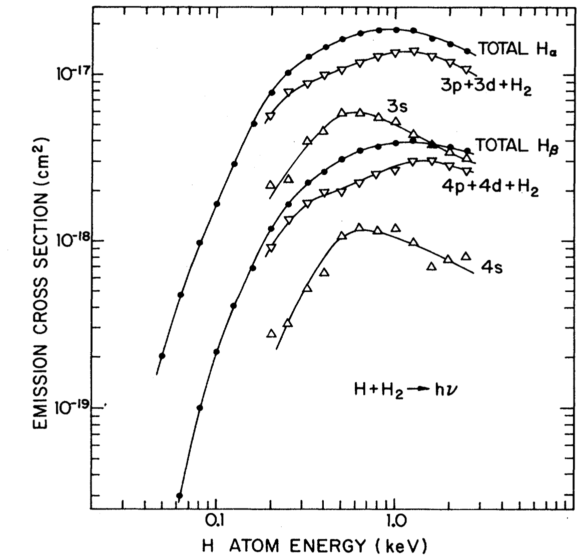

FIG. 3. Total H_α and H_β emission cross sections for the $H+H_2$ reaction. Contributions to these emission cross sections from decay of the hydrogenic states listed were separated for H-atom energies above 0.2 keV. Contribution labeled H_2 results from dissociative excitation of the target H_2 molecules.

states. (Recall that radiation from decay of the *ns* states of hydrogen must be unpolarized, because of their spherical symmetry. Thus the two sets of values are related by the fractions of the radiations resulting from decay of the *ns* and the *np* + *nd* states populated during the collisions.) The uncertainties associated with these data are indicated by the flags on some of the points plotted.

The emission cross section and radiation polarization data for $H+H_2$ collisions are shown in Figs. 3 and 4, respectively. The emission cross sections labeled $np+nd+H_2$ reflect the inability to resolve those parts of the H_α and H_β emissions resulting from decay of the $np+nd$ excited states of the projectile H atoms from those parts resulting from dissociative excitation of the target H_2 to all available Balmer-line-emitting states. Because of this situation, and the fact that the contributions to the total H_α and H_β signals resulting from decay of the excited *ns* states of the projectile H atoms are relatively small, only the polarizations of the total emitted radiations are presented in Fig. 4.

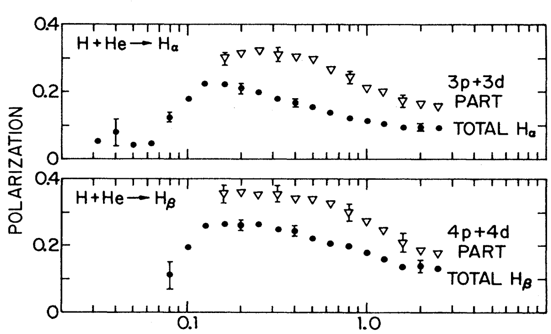

FIG. 2. Polarizations of the total (solid circles) and $np+nd$ parts (open triangles) of the H_α and H_β emissions resulting from H+He collisions.

While the uncertainties in the H_α and H_β emission cross sections from the various *nl* states of the projectile H atoms excited in $H+H_2$ collisions should be comparable to those cited above for H + He collisions, the uncertainty analysis is complicated by the H_α and H_β emission from dissociative excitation of the target H_2. Because the target hydrogen atoms excited to the longer-lived 3*s* and 4*s* states by dissociative excitation of H_2 will have some kinetic energy (from their separation along a repulsive potential-energy curve as well as from momentum transfer from the projectile H atoms during the collision), some of these excited atoms, if suitably directed, could move to positions outside the viewing field of the photon detector before decaying. While the energy and angular distributions of these excited dissociation fragments would both have to be known (which they are not) to determine properly the fractions of the total H_α and H_β emissions not detected for this reason, crude estimates suggest that it is unlikely that more than 5% of the emitted photons from this source should escape detection. Because the contributions to the

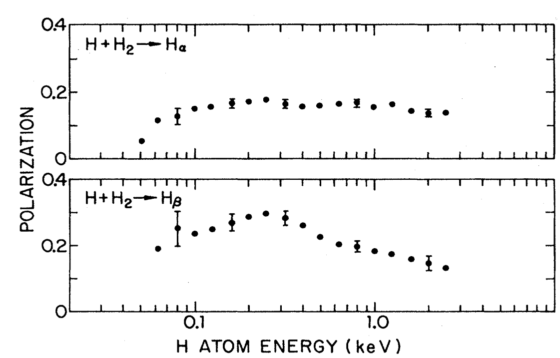

FIG. 4. Polarizations of the total H_α and H_β emissions resulting from $H+H_2$ collisions.

total H_α and H_β signals from dissociative excitation of H_2 are probably not large (as will be discussed below), the uncertainties in the emission cross section data shown in Fig. 3 should not be significantly larger than those cited for H + He collisions.

The only other experimental data which the present results can be compared with are those of Williams *et al.*,[5] who investigated H_α emission from $H + H_2$ collisions for H-atom energies down to 2 keV. Where the two sets of data overlap, their agreement is very satisfactory, being well within mutual uncertainties. These workers were also able to separate the H_α emissions resulting from dissociative excitation of H_2 from those resulting from excitation of the projectile H atoms (by observation of the Doppler shift of the emitted radiations). While the fraction of the total H_α emission from dissociative excitation of H_2 was found to be 26% for an H-atom energy of 10 keV, it decreased to 17% at 5 keV, and to only 11% at 2 keV. Based upon this trend, the present authors would be surprised if H_α emission from dissociative excitation of H_2 should emerge as a major source of the here-observed emission at lower H-atom energies.

Hughes *et al.*[6] have also measured the cross sections for H_α emission from excited projectile H atoms for both H + He and $H + H_2$ collisions for H-atom energies between 10 and 35 keV. In this energy range, their results for $H + H_2$ collisions lie about a factor of 2 below the emission cross-section data of Williams *et al.*[5] Based upon the good agreement between the present results and those of Williams *et al.*,[5] it seems probable that the emission cross-section values reported by Hughes *et al.*[6] for the $H + H_2$ reaction are too low, thus calling into question their data for the case of H + He collisions as well.

As noted in Sec. I, the total H_α emission cross section for $H^+ + He$ collisions was also approximately determined for H^+ energies of 2.0 and 1.25 keV. The cross-section values obtained were 1.7 and 0.8×10^{-20} cm^2 at these H^+ energies, respectively, with uncertainties of about $\pm 50\%$ of the values cited. Because of the very small magnitude of this cross section, and its decreasing value with decreasing H^+ energy, experimental limitations prevented extension of these measurements to lower H^+ energies (or to the case of H_β emissions from the reaction).

These limitations centered about the fact that, for H^+ energies below 1 keV, the H_α emission cross section for $H^+ + He$ collisions was more than four orders of magnitude smaller than the H_α emission cross section(s) for H^+ impact on the residual atoms and/or molecules (of undetermined species) present in the target cell as a background gas. Thus, even with He to background-gas pressure ratios of 10^4 or so in the target cell, the measured H_α signal was still dominated by H^+ impact on the background-gas species. While this problem could be overcome somewhat by operating the target-cell He pressure at values in excess of 10^{-3} Torr, second-collision effects became overwhelming in this higher-pressure region. Because the H_α emission cross section for H + He collisions is also much larger than that for $H^+ + He$ collisions (by about three orders of magnitude for projectile energies in the 1-keV range), conversion of only a very small fraction of the H^+ traversing the target cell into fast H atoms via electron-capture reactions can result, from the subsequent H + He collisions, in greatly increased H_α signals. In fact, even for the higher H^+ energies at which the H_α emission cross section was approximately determined here (by detailed studies of the He target-cell-pressure dependence of the H_α signals), ignoring the above-mentioned problems would have yielded "apparent" cross-section values much larger than those cited above.

While there are no other measurements with which these results can be directly compared, the data of Dawson and Loyd[7] do extend down to an H^+ energy of 3.2 keV, where the total H_α emission cross section for $H^+ + He$ collisions was reported to be 3.1×10^{-19} cm^2. However, this value is almost a factor of 20 larger than the value of 1.7×10^{-20} cm^2 obtained here for an H^+ energy of 2.0 keV, and so rapid a variation of this cross section with H^+ energy is highly improbable. These sets of data thus appear to be in conflict by approximately an order of magnitude in the 2—3-keV range of H^+ energies.

III. DISCUSSION

While the H_α and H_β emission cross sections for H + He and $H + H_2$ collisions are of interest in themselves for a variety of applications, of more fundamental interest are the cross sections for excitation to the various *nl* states of hydrogen which decay via these emissions. Even though the cross sections for excitation to all these *nl* states cannot be accurately determined from the emission cross-section data in Figs. 1 and 3, a considerable amount of information can be obtained from these results.

For example, because high-lying *np* states of hydrogen decay highly preferentially to the 1*s* and 2*s* states, cascade population of the 3*s* and 4*s* states from such high-lying *np* states (via Paschen- and Brackett-series transitions) excited during the collisions of interest should not represent significant sources of H_α and H_β emission. Thus, because the branching ratio for the $3s \to 2p$ hydrogenic transition is 1.00, the cross section for excitation to the 3*s* state should be quite close to that for H_α emission from decay of this state. Similarly, the cross section for excitation to the 4*s* state should be about 1.71 times as large as that for H_β emission from this state, as a result of the $4s \to 2p$ branching ratio of 0.584.

A further consequence of the preference of excited *np* states of hydrogen to decay to the 1*s* state (via Lyman-series emissions) is the fact that the branching ratios for the $3p \to 2s$ and $4p \to 2s$ Balmer-line-emitting transitions are only about 0.12. In contrast, the branching ratios for the $3d \to 2p$ and $4d \to 2p$ transitions have the much larger values of 1.00 and 0.746, respectively. Thus, unless the excitation cross sections for the 3*p* and 4*p* states of hydrogen are much larger than those for the 3*d* and 4*d* states, the bulk of the H_α and H_β emission cross sections labeled $np + nd$ in Figs. 1 and 3 should result from *nd*-state decay.[8] The cross section for excitation to the 3*d* state of hydrogen in H + He collisions should thus not be substantially below the H_α emission cross section labeled $3p + 3d$ in Fig. 1, and that for excitation to the 4*d* state should be a reasonable fraction of 1.34 times the H_β emission cross section labeled $4p + 4d$. Similar arguments can be made for the case of $H + H_2$ collisions, to the extent that H_α and H_β emissions from dissociative excitation of H_2 can be ignored.

The above arguments assume that cascade population of the $3d$ and $4d$ states of hydrogen can be ignored. While the assumption should be reasonably valid for transitions from higher-lying np states (because of the small branching ratios for these transitions), no similar conclusions can be reached about possible $nf \rightarrow 3d$ and $nf \rightarrow 4d$ transitions (which have large branching ratios). Because the excitation cross sections for these nf states are not known, the only kinds of arguments that can be made against the likelihood of such cascade effects are similar to those discussed in the earlier report on H+Ar collisions,[1] having to do with how such effects would perturb the measured increase in photon signal as a function of distance into the target cell. While these kinds of arguments are valid, they are not highly definitive, demonstrating only that these cascade effects are not major sources of the observed H_α and H_β emissions.

It is interesting to note that the cross sections for excitation of projectile H atoms to the $3s$ and $4s$ states in H+He and $H+H_2$ collisions have very similar magnitudes and H-atom—energy dependences, in spite of the considerable differences between the internal structures of these target species. Even more remarkable, perhaps, is the fact that the H-atom—energy dependences of these ns-state excitation cross sections are also very similar to those reported earlier[1,2] for H-atom impact on Ar, N_2, and O_2, which also reach their maximum values for H-atom energies between 0.5 and 1 keV. (Curiously, the ns-state excitation cross sections for H impact on these heavier targets are also quite similar in magnitude, averaging about four times as large as those for He and H_2 targets. Note the approximate scaling of all these cross-section magnitudes with the number of outer-shell electrons present in these various target species.)

It is also interesting that the ratio of the cross sections for excitation to the $4s$ and $3s$ states of hydrogen is about 0.33 for both H+He and $H+H_2$ collisions. Similar ratios, although slightly smaller, were also obtained for H-impact collisions with Ar, H_2, and O_2. This ratio is not far below the value of 0.42 predicted by the n^{-3} scaling law for ns-state excitations of hydrogen (which scaling might be expected to be valid for much higher H-impact energies in such collisions, or for electron-impact excitation of hydrogen atoms).

The surprising consistency of these ns-state excitation cross sections for all the reactions discussed above seems to point to an excitation mechanism which is substantially independent of the details of the interactions involved. Could it be, for example, that these excitation processes can be thought of simply as excitation of the projectile H atoms by the orbital electrons of the target particles? While such an interpretation must be considered to be rather naive, it is consistent with the experimental observations. In any case, it would appear that whatever the operative mechanism, it is not strongly dependent on the nature of the rather different transient molecular complexes which must exist during such diverse interactions.

In sharp contrast to the similarities in all the ns-state excitation cross sections discussed above are those for H_α and H_β emission from decay of the $np+nd$ states of the collisionally excited projectile H atoms. For example, for the H+Ar reaction,[1] the H_α emission cross section (presumably resulting largely from decay of the $3d$ state of hydrogen) reaches its maximum for an H-atom energy of about 0.07 keV, where its absolute value is about two orders of magnitude larger than that for H+He collisions at a comparable H-atom energy. While the H_α emission cross section for H+He collisions does exhibit a structure in the H-atom—energy range below 0.1 keV, the polarization of the H_α emission from this reaction is here quite small, in contrast to the larger value (~ 0.3) resulting from H+Ar collisions. Thus, both the cross-section magnitudes and radiation polarizations point to significant differences in the details of the excitation mechanisms involved in these collisions at low H-atom energies. Essentially identical arguments can be made for the case of H_β emissions from these reactions.

For the case of H+Ar collisions, it was suggested[1] that the large cross sections for nd-state excitation at low H-atom energies, and the polarizations of the consequent H_α and H_β radiations, could be explained by the existence of a transient ionic state during the interaction; i.e., by thinking of the reaction sequence $H + Ar \rightarrow H^- + Ar^+ \rightarrow H^* + Ar$. The nd-state—excited hydrogen atoms would thus be produced on the outward-bound leg of the collision where, at fairly large internuclear separations, the potential-energy curve for the ionic state will cross (at least in a diabatic sense) the curves leading to the postcollision H^*+Ar products. However, for the case of H+He collisions, no such crossings are possible, because of the high potential energy of the H^-+He^+ state (resulting from the large ionization potential of He). Thus the absence of a large maximum in the H_α and H_β emission cross sections for H+He collisions at low H-atom energies is certainly not inconsistent with this model of the H+Ar interaction.

Comparisons of the present H_α and H_β emission cross-section data for $H+H_2$ collisions with those obtained earlier for molecular N_2 and O_2 targets[2] also point out some interesting differences. For example, for H-atom energies above about 0.3 keV, the H_α emission cross sections for H impact on N_2 and O_2 are dominated by decay of the $3s$-state—excited projectile H atoms, while for H_2 targets, the bulk of the observed emission probably comes from decay of the $3d$ state. In addition, the polarization of the H_α radiation from $H+H_2$ collisions is fairly large and positive, in contrast to the generally smaller and negative polarizations for $H+N_2$ and $H+O_2$ collisions in this range of H-atom energies.

As discussed earlier for H+Ar collisions,[1] the polarizations of the H_α and H_β emissions resulting from decay of the $3d$-state—excited projectile H atoms would be $+0.48$, $+0.26$, and -0.70 for decay of the pure $m_l=0$, ± 1, and ± 2 sublevels, respectively, for that interaction. While interpretation of such polarization data may depend on the nature of the interaction (i.e., at what internuclear separations in the rotating reference frame of the collision the excited hydrogen atoms are produced), it appears likely that projectile H atoms excited to the $m_l=0$ and ± 1 sublevels of the $3d$ state in H+He collisions should make major contributions to the H_α and H_β radiations, because they are observed to be strongly positively polarized for projectile H-atom energies above 0.1 keV. For the cases of the molecular targets discussed immediately above, the differences in the H_α polarizations noted also seem to imply that differing m_l sublevel populations are being produced in the collisions, although the issue is here somewhat complicated by the basically triatomic nature of these interactions (i.e., by the uncertainty in correlating a "molecular axis" for the collisions with the H-atom-beam axis).

In summary, the H_α and H_β emission cross sections reported here can be used to determine some of the cross sections for excitation to the various *nl* states of the projectile H atoms populated in H+He and $H+H_2$ collisions. The cross sections for excitation to the 3*s* and 4*s* states of hydrogen in these collisions are remarkably similar to those obtained earlier[1,2] for the cases of H impact on Ar, N_2, and O_2, while those for excitation to the 3*d* and 4*d* states are highly dependent on the particular target particle involved. The planned extension of this work to include Lyman-α emission cross-section measurements and to determinations of the 2*s*-state excitation cross sections in such collisions should provide further insight into the interaction mechanisms involved in the excitation processes.

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[8]If the H_α emission cross section labeled $3p+3d$ in Fig. 1 were to result entirely from the $3p \rightarrow 2s$ hydrogenic transition, the 3*p*-state excitation cross section would have to be about 3.5×10^{-17} cm^2 at 1-keV H-atom energy. This is considered unlikely, because the 2*p*-state excitation cross section for H+He collisions appears to be only slightly larger than this value, as determined from the Lyman-α emission cross-section measurements of J. H. Birely and R. J. McNeal, Phys. Rev. A 5, 257 (1972).