

used in a number of papers published later by various authors.^{2,5}

We would like to take this opportunity to point out that the comment of Bassel and Gerjuoy⁵ that the wrong matrix element is used in reference 1 to evaluate the capture amplitude ignores the fact that in reference 1 the matrix element evaluated has been proved to be approximately equal to the correct matrix element and

⁵ R. H. Bassel and E. Gerjuoy, *Phys. Rev.* **117**, 749 (1960); M. R. C. McDowell, *Proc. Roy. Soc. (London)* **A264**, 277 (1961).

that at very high energy, where the Born approximation can be taken to be exact, the equality of these two matrix elements is exact. It is, therefore, difficult to see how Drisko's estimates referred to by Bassel and Gerjuoy can indicate that the error caused by the use of the "wrong" matrix element is serious in the high-energy limit. Our belief, which is based on the proof given in reference 1, is that the matrix element evaluated by Pradhan and used by us in the present work for the computation of the total cross section is so close to the correct one that the error is negligible.

Balmer Emissions Induced by Proton Impact on Molecular Hydrogen*

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Absolute cross sections for the production of H_{α} , H_{β} , and H_{γ} emissions by proton impact on molecular hydrogen have been measured. Emissions produced through the process of electron capture into excited states by fast protons are Doppler shifted from emissions produced through dissociative excitation of the target gas, which allows separate measurements of these processes. Comparisons are made with theoretical calculations of proton impact on atomic hydrogen.

I. APPARATUS

A POSITIVE-ION accelerator has been built at the University of Arkansas to accelerate ions through a maximum potential of about 140 kV for the purpose of studying the spectra induced by ion impact on gases. The ion beam is magnetically analyzed as it is bent through 30° into the collision chamber. Figure 1 shows the details of the differentially pumped collision cham-

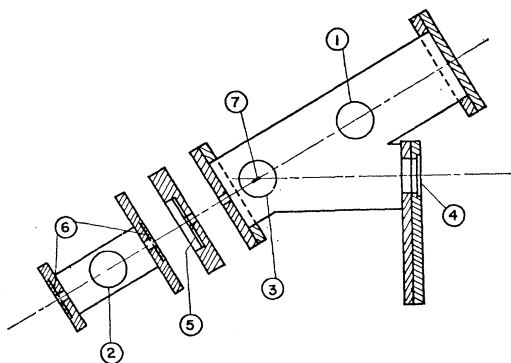


FIG. 1. Collision chamber—(1) gas inlet, (2) differential pumping outlet, (3) McLeod gauge, (4) view port, (5) electron repeller, imbedded in Lucite which insulates the collision chamber, (6) collimating apertures ($\frac{1}{16}$ -in. holes), (7) Pirani gauge.

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ber. Not shown is a liquid-air trap at the end of the collision chamber. This trap was installed to remove condensable vapors from the collision chamber.

Spectroscopic observation of the collision region is made at an angle of 30° to the beam. This allows measurements on Doppler-shifted emissions produced through the process of electron capture into excited states by fast protons to be separated from the unshifted radiation produced by direct excitation processes in the target gas. A JaCo 500 mm Ebert spectrometer was calibrated for use in the $\lambda 3800$ to $\lambda 6600$ Å spectral range. The calibration procedure has been previously described.¹ The spectrometer now uses an EMI 6095B photomultiplier as a detector.

Pressure measurements are made with a trapped McLeod gauge while a Pirani gauge is used to monitor the pressure. The hydrogen was introduced into the collision chamber via a heated palladium leak. Pressure ranged from 1.5μ Hg for the low-energy work to 9μ for the higher energies.

II. RESULTS AND DISCUSSION

Balmer radiations, H_{α} , H_{β} , and H_{γ} were measured for proton impact on H_2 . These emissions were linear with current and above 10 keV they were measured in a pressure range where the emissions were linear with pressure. Below 20 keV, we suffer a loss in beam current

¹ R. H. Hughes, R. C. Waring, and C. Y. Fan, *Phys. Rev.* **122**, 525 (1961).

that limits our accuracy. Particularly bad are the 5-keV points where we found it difficult to ascertain for certain whether or not we were in a pressure range where the emissions are linear with pressure. Above 20 keV our absolute measurements should be good to within about 40%. The relative measurements, however, should be better.

Our definition of cross section follows from the equation $n = \sigma \rho F$, where n is the number of photons emitted from a cubic centimeter, σ is the cross section, ρ is the molecular density in the chamber, and F is the proton flux. In order to compare the results with calculations on atomic hydrogen we do at times refer to a cross section per hydrogen atom, but in these cases the cross section is always specified as such.

Our results are displayed in Fig. 2 for H_α and H_β

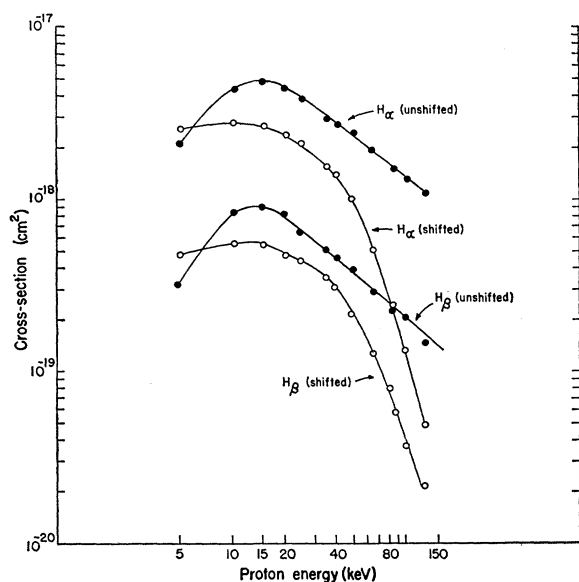


FIG. 2. H_α and H_β emissions produced by proton impact on H₂.

emissions, both Doppler shifted and unshifted. H_γ cross sections are shown in Figs. 3 and 6.

A. The Doppler-Shifted Emissions

These emission cross sections are displayed in Fig. 3. Presumably these emissions are produced through the decay of fast hydrogen atoms resulting from the electron capture from hydrogen by the incident protons according to the most probable transfer reaction: $H^+ + H_2 \rightarrow H^* + H_2^+$. These emissions appear to peak at about 10 keV. A two-step process is possible where the proton captures an electron and the resulting hydrogen atom is excited by a second collision. In the range 5–10 keV (where charge transfer is a maximum) and at a pressure of 1.5μ Hg the mean free path for charge transfer in hydrogen is about 25 cm. We observe the beam just as it enters the collision chamber through a pumped

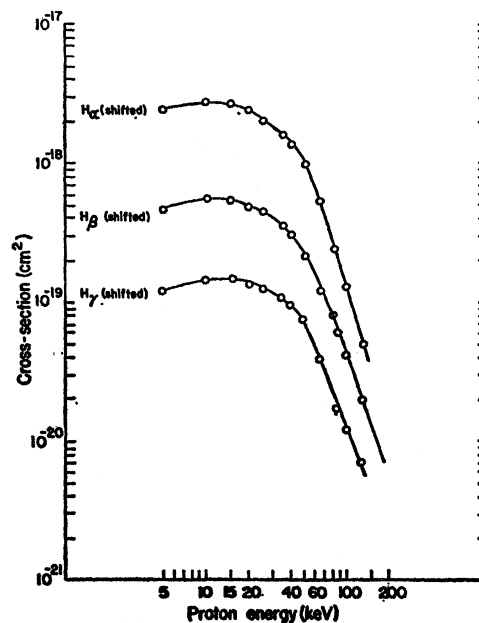


FIG. 3. Balmer emission cross sections from proton capture of an electron from molecular hydrogen.

chamber. The viewing region itself is about 3 cm long. Thus, the two-step process would seem somewhat rare. The linearity of the emissions with pressure attests to the single-collision event of electron capture into excited states. (As previously pointed out, the certainty

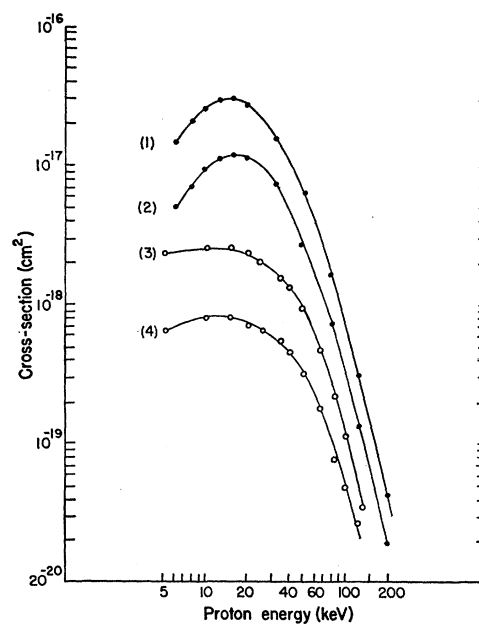


FIG. 4. Cross sections for populating the $n=3$ and $n=4$ levels of fast hydrogen atoms through electron capture from hydrogen by protons. (1) Theory, $n=3$, H^+ on H (Bates and Dalgarno); (2) theory, $n=4$, H^+ on H (Bates and Dalgarno); (3) experimental estimate, $n=3$, H^+ on H₂ per H atom; (4) experimental estimate, $n=4$, H^+ on H₂ per H atom.

of this last statement can be questioned for the 5-keV points.)

Bates and Dalgarno² have calculated electron capture into excited states by protons from atomic hydrogen using the first Born approximation. They calculate that excited-state capture should pass through maxima at about 15 keV. Our apparent peaks occur at about 10 keV for proton impact on molecular hydrogen. Figure 4 displays level cross sections calculated by Bates and Dalgarno together with our own experimental estimates of the level cross sections per hydrogen atom. We estimated the electron capture cross section into the $n=3$ and $n=4$ levels from the Doppler-shifted H_α and H_β line cross sections. The factor required to change the line cross section to level cross section can be derived easily (neglecting cascade). For example, consider excitation to the $n=3$ level. Let $dN(3s)/dt = \sigma(3s)\rho F$ be the rate at which the $3s$ level is being populated by proton impact; $N(3s)$ is the number of atoms per cm^3 being placed in the $3s$ level, $\sigma(3s)$ is the level cross section, ρ the target gas density, and F is the proton

flux. Similar equations will hold for the $3p$ and $3d$ levels. Thus, $\sigma(n=3) = \sigma(3s) + \sigma(3p) + \sigma(3d)$. The rate at which the $3s$ level is depopulated by radiative processes is $dN(3s)/dt = N(3s)/T_{3s}$, where T_{3s} is the mean radiative lifetime of the $3s$ state. In equilibrium, $N(3s) = T_{3s}\sigma(3s)\rho F$ with similar equations holding for the $3p$ and $3d$ levels. The rate at which H_α photons are being emitted,

$$n(H_\alpha), \text{ is } n(H_\alpha) = \sigma(H_\alpha)\rho F = N(3s)A(3s \rightarrow 2p) \\ + N(3p)A(3p \rightarrow 2s) + N(3d)A(3d \rightarrow 2p),$$

where the A 's are the indicated transition probabilities. Substituting, we find

$$\sigma(n=3) = \sigma(H_\alpha) \\ \times \left[\frac{1 + R_1 + R_2}{1 + R_1 T_{3p} A(3p \rightarrow 2s) + R_2 T_{3d} A(3d \rightarrow 2p)} \right], \quad (1)$$

where $R_1 = \sigma(3p)/\sigma(3s)$ and $R_2 = \sigma(3d)/\sigma(3s)$. For $n=4$ we have

$$\sigma(n=4) = \sigma(H_\beta) \left[\frac{1 + R_3 + R_4}{T_{4s} A(4s \rightarrow 2p) + R_3 T_{4p} A(4p \rightarrow 2s) + R_4 T_{4d} A(4d \rightarrow 2p)} \right], \quad (2)$$

where $R_3 = \sigma(4p)/\sigma(4s)$ and $R_4 = \sigma(4d)/\sigma(4s)$. We neglect $\sigma(4f)$ which is likely to be small. [An erroneous equation similar to (2) was published in a previous paper.³]

The cross-section ratios were obtained from Bates and Dalgarno. The factor required to change the line cross sections to level cross sections is not too sensitive to these ratios, at least within the limits of reasonable ratios. If we had chosen Mapleton's calculations⁴ for H^+ on He to obtain these ratios, it would have made a maximum difference of about 15% in the multiplying factor for energies greater than 20 keV. At the higher energies (greater than 100 keV) the cross-section ratios themselves are not particularly sensitive to which calculation is chosen.

The value of a comparison of our work with the Born approximation calculations of proton impact on atomic hydrogen can be questioned on the grounds that (1) the Born approximation at low impact velocities is quite poor and (2) the assumption that the hydrogen molecule can be treated as equivalent to two hydrogen atoms may not be particularly valid.⁵ The comparisons in this paper are, therefore, presented in the spirit of academic interest rather than as a very serious attempt at comparison with theory.

² D. R. Bates and A. Dalgarno, Proc. Phys. Soc. (London) **A66**, 972 (1953).

³ R. H. Hughes, J. L. Philpot, and C. Y. Fan, Phys. Rev. **123**, 2084 (1961).

⁴ R. A. Mapleton, Phys. Rev. **122**, 528 (1961).

⁵ T. F. Tuan and E. Gerjuoy, Phys. Rev. **117**, 756 (1960).

We also calculated the fraction of total capture that results in Doppler-shifted H_α , H_β , H_γ emission, using the total capture cross sections tabulated by Allison.⁶ These fractions are displayed in Fig. 5. Bates and Dalgarno calculated this fractional quantity for H_α and H_β . Although our fraction of capture resulting in H_α emission peaked at roughly their predicted energy, our experimental fractions were about a factor of 10 less.

B. The Doppler-Unshifted Radiation

Cross sections for the production of these radiations are displayed in Fig. 6.

Three excitation mechanisms might be possible:

- (a) $H^+ + H_2 \rightarrow H + H^* + H^+$
- (b) $H^+ + H_2 \rightarrow H^+ + H^* + H^+ + e$
- (c) $H^+ + H_2 \rightarrow H^+ + H^* + H$

The maximum excitation of the unshifted Balmer lines occurs at about 15 keV. At this energy, processes (a) and (b) can be ruled out by Keene's⁷ failure to observe an appreciable number of slow protons in his study of proton impact on H_2 . This leaves simultaneous dissociation and excitation (c) as the most probable mechanism. Bates and Griffing⁸ have calculated the

⁶ S. K. Allison, Rev. Mod. Phys. **36**, 1137 (1958).

⁷ J. P. Keene, Phil. Mag. **40**, 369 (1949).

⁸ D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **A66**, 961 (1953).

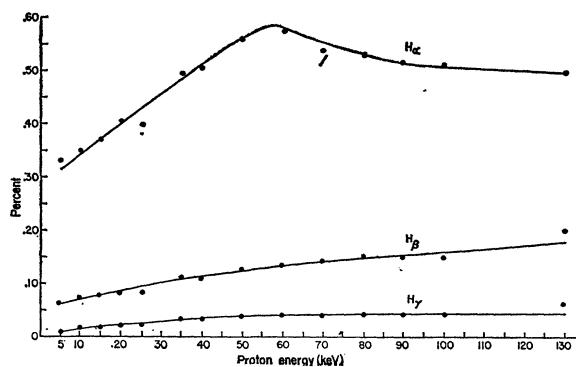


FIG. 5. Fraction of total charge transfer resulting in Doppler-shifted H_α, H_β, and H_γ emissions in H₂.

population of excited states in the reaction $H^+ + H \rightarrow H^+ + H(n)$ for $n=2, 3$, using the first Born approximation. We have included our interpretation of their results in Fig. 6 for $n=3$. We also included our estimate of the population cross section of the $n=3$ level by proton impact on molecular hydrogen per hydrogen atom. We referred to the calculations of Bates and Griffing to obtain the factors required to transform our H_α measurements to level measurements.

The unshifted Balmer emissions seem to go roughly as $E^{-0.85}$ where E is the proton energy. This seems to hold from about 25 keV to the higher energies.

Bates^{9,10} has done further work on the problem $H^+ + H(1s) \rightarrow H^+ + H(n=2)$ and has found that distortion strongly influences the calculation of $n=2$ population, particularly the $2s$ level. This procedure succeeds in lowering the theoretical value at 10 keV by about a factor of 3.6. Also, the relative reduction in s state population would increase the factor by which we must multiply our line cross section to obtain the level cross section to the extent that fair agreement can be obtained at 10 keV. However, the inclusion of distortion in the calculations makes the level population peak at about 35 keV. Further refinements in the theory, such as inclusion of rotation coupling and back coupling,

⁹ D. R. Bates, Proc. Phys. Soc. (London) **73**, 227 (1959).

¹⁰ D. R. Bates, Proc. Phys. Soc. (London) **77**, 59 (1961).

make little difference, at least in the $2p$ level.¹¹ Actually the Born approximation seems to describe the general shape of our curve fairly well.

In closing it is of some interest to compare our results with the recent Lyman alpha study by Dunn *et al.*¹² Extrapolating their results from 3 to 5 keV seems to indicate that the ratio of the H_α total radiation (shifted plus unshifted) to Ly_α total radiation at 5 keV is about 15%.

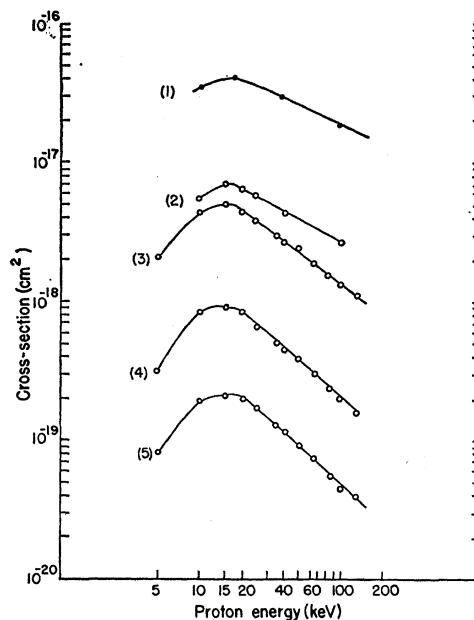


FIG. 6. Excitation cross sections for proton impact on hydrogen. (1) Theory, $n=3$ level, H^+ on H (Bates and Griffing); (2) experimental estimate, $n=3$ level, H^+ on H_2 per H atom; and (3) H_α; (4) H_β; (5) H_γ emissions from H^+ on H_2 .

ACKNOWLEDGMENTS

The authors wish to thank William Evans, David Kay, and Darrell Bolding for their aid in obtaining data.

¹¹ R. J. Bell and B. G. Skinner, Proc. Phys. Soc. (London) **80**, 404 (1962).

¹² G. Dunn, R. Geballe, and D. Pretzer, Phys. Rev. **128**, 2200 (1962).