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CROSS SECTIONS FOR THE DESTRUCTION OF 1- TO 25-keV-PER-NUCLEON $x^1\Sigma_g^+$ AND $c^3\pi_u$ HYDROGEN MOLECULES IN COLLISIONS WITH H_2 GAS *

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January 1971

Measurements of the cross sections for the total loss and the ionization of $\chi^1\Sigma_g^+$ H₂ [1 \leq E (keV/nucleon) \leq 25] and $c^3\Pi_u$ H₂ molecules [3 \leq E (keV/nucleon) \leq 20] in collisons with H₂ gas are reported. The $c^3\Pi_u$ molecules were formed by electron capture by H₂⁺ ions in Mg vapor. The total-loss cross section of the $c^3\Pi_u$ state is about 5 times larger than the total-loss cross section of the $\chi^1\Sigma_g^+$ state.

As part of a study of methods of forming fast neutral hydrogen beams for injection in controlled thermonuclear experiments, Hiskes showed that beams of hydrogen molecules in the (1 so2pm) c $^3\Pi_u$ electronic state would be desireable for injection into plasma targets with densities in the 10^{11} to 10^{14} cm $^{-3}$ range. Charge transfer collisions of H_2^+ with alkali or alkaline-earth targets are expected to lead to relatively large populations of this state. The calculated radiative lifetimes of all vibrational levels of this state are long ($\gtrsim 10^{-4}$ sec), at least for homonuclear molecules, compared with transit times in experiments of interest, and the transition rates are not appreciably quenched by electric fields less than about 10^4 V/cm 3

Experimentally, we find that a large fraction of the molecules formed

by electron capture in a thin magnesium vapor target are indeed in an excited state, e.g., about 40% for incident 20-keV $\rm H_2^{-1}$ ions. (The fraction of the incident ion beam that can be converted to excited molecules has not yet been determined.) These large fractional yields make it possible to measure collisional-loss cross sections of two classes of $\rm H_2$ molecules, which we presume are in the $\rm c^3\Pi_u$ state and the $\rm X^1\Sigma_g^{-1}$ (ground) state. No significant fraction ($\lesssim 5\%$) of excited molecules was observed for electron capture in $\rm H_2$ or $\rm N_2$ gas, in agreement with predictions. The measurements reported here are loss cross sections in $\rm H_2$ gas. We have made measurements with incident $\rm H_2^{-1}$, $\rm D_2^{-1}$, and $\rm HD^{+1}$ ion beams. When the cross sections are compared at the same projectile velocity, the results are the same within the experimental uncertainties; hence, in the remainder of the paper we treat all projectiles as if they were $\rm H_2^{-1}$ ions or $\rm H_2$ molecules.

The experimental method used for the analysis of excited $\rm H_2$ molecules is similar to that introduced by Gilbody et al. for metastable He atoms. The energetic $\rm H_2$ molecules were produced by electron capture when a momentum-analyzed beam of $\rm H_2^{-1}$ ions traversed a neutralizing cell of Mg vapor, or $\rm H_2$ or $\rm N_2$ gas. The emerging charged particles were swept out of the beam with an electric field, and the $\rm H_2^{-1}$ component, detected with a Faraday cup, provided a monitor of the $\rm H_2$ beam intensity. The neutral beam, consisting of ground-state and excited H and $\rm H_2$, was analyzed by measuring the attenuation of $\rm H_2$ in a target cell containing $\rm H_2$ gas. To insure that the exiting $\rm H_2$ beam reflected only attenuation, without the contribution of two-step processes, a transverse electric field of about 8 kV/cm in the target cell was used to deflect charged particles, produced

by collisions in the cell, immediately after formation.

Two methods were used for particle detection: For method I we employed particle-counting techniques. The particles were detected with a CsI (Tl) crystal mounted on a photomultiplier tube. Energetic H_2 molecules were distinguished from two separate H atoms, which gave the same pulse height, by placing a low-transparency (\approx 1%) mesh in front of the scintillator. The mesh discriminated against 2H events because the atoms are spatially separated when they arrive at the mesh. Method I is limited to energies greater than about 20 keV; below this energy it is difficult to resolve the single pulse heights owing to the atomic portion of the beam from the double pulse heights of the molecular portion.

For method II we used a third gas cell held at a constant pressure to strip the fast molecules of an electron, and measured the resulting H₂⁺ current with a Faraday cup. This method yields less information than method I but is applicable to cross section measurements below 20 keV.

A semilogarithmic plot of the fraction of the H_2 beam transmitted through the target cell vs target thickness is shown in Fig. 1. The solid curve shows the attenuation of the total H_2 beam. If we assume that there are two beam components of interest (background is negligible), this curve can be unfolded into two exponential curves (A and B) by extrapolating the straight-line portion (A) to zero target thickness and taking the difference (B) between this extrapolation and the total attenuation. The slope of A yields the total-loss cross section for the $X^1\Sigma_g^+$ state; the slope of B yields the total-loss cross section for the long-lived ($\gtrsim 10^{-6}$ sec) excited state, which we assume is the $c^3\Pi_u$ state. With method I the intercept of B gives the fraction of the beam in the $c^3\Pi_u$ state; this will be discussed in a future paper.

One of the cross sections contributing to the total-loss cross section, that for ionization $(H_2 \to H_2^{-1})$, was also determined in the present experiment. For this measurement the transverse electric field in the target cell was turned off, and the H_2^{-1} component was measured as a function of target thickness, Π , up to $\Pi = 2 \times 10^{14}$ molecules/cm². When the incoming H_2 beam contained no observable fraction of $c^3\Pi_u$ states (e.g., when an H_2 or N_2 neutralizer was used) the linear relation between H_2^{-1} and Π yielded σ_1 , the cross section for ionization of the ground state. When the incident H_2 beam did contain an observable fraction of $c^3\Pi_u$ molecules (thin Mg neutralizer), the same technique yielded a composite cross section $(1-f)\sigma_1^- + f\sigma_1^{-*}$ (approximately twice σ_1^-), where f is the fraction of the beam in the $c^3\Pi_u^-$ state, obtained from Fig. 1 for the same neutralizer condition. Since σ_1^- and f were determined independently, the cross section for ionization of the $c^3\Pi_u^-$ state, σ_1^{-*} , can be determined.

Preliminary results for the energy variation of the total-loss and ionization cross sections for excited and ground state $\rm H_2$ molecules colliding with $\rm H_2$ gas are shown in Fig. 2.

To check the possibility of a systematic error, we measured the well-known latotal-loss cross section for atomic hydrogen passing through $\rm H_2$ gas. Our measurements agreed with those of Stier and Barnett to within $\pm 5\%$. The present attenuation technique for measuring excited-state collisional cross sections has yielded results that we believe are accurate to about $\pm 25\%$. The major portion of this uncertainty, estimated from the

reproducibility of data points, is due to fluctuations in the beam position in the apparatus that affect the monitoring of the primary beam. Uncertainties in target thickness and other quantities are negligible in comparison. The uncertainty associated with the ground state measurements is estimated to be about $\pm 10\%$.

Comparison of the data for the $X^1\Sigma_g^+$ H₂ ionization cross section in H₂ gas shows excellent agreement with the results of McClure⁷ (see Fig. 2). McClure has also used a very different technique to measure the totalloss cross section of $X^1\Sigma_g^+$ molecules at an energy of 10 keV: again, good agreement with the present measurements is found in Fig. 2. We have not found any data on collisional cross sections for the $c^3\pi_u$ state to compare with present results.

To investigate the $c^3\Pi_u$ state for heteronuclear molecules we used ${\rm HD}^+$ as a fast projectile. Within experimental uncertainties the results for HD are identical with the results for H₂ and D₂, indicating that the lifetimes in the case of HD ($c^3\Pi_u$) are larger than the time of flight of the projectile ($\gtrsim 10^{-6}$ sec) in the present experiment.

We are indebted to Dr. J. R. Hiskes for bringing this problem to our attention and for many useful discussions.

FOOTNOTES AND REFERENCES

- *Work done under the auspices of the U. S. Atomic Energy Commission.
- 1. J. R. Hiskes, Bull. Am. Phys. Soc. 13, 308 (1968).
- 2. J. R. Hiskes, Phys. Rev. 180, 146 (1969).
- 3. R. P. Freis and J. R. Hiskes, Phys. Rev. A2, 573 (1970).
- 4. H. B. Gilbody, R. Browning, and G. Levy, J. Phys. B 1, 230 (1968).
- 5. This assumption is based on the following argument: We observe a component in the H, beam that has a loss cross section larger than that for the ground state. We assume this is an excited state or The time of flight from the neutralizer cell is on the order of 10⁻⁶ sec. If we search for excited states with lifetimes of this order or longer, we find that the $c^3\Pi_{u}$ electronic state³ [W. Lichten, Phys. Rev. 120, 848 (1960)] and the hydrogen-like Rydberg states with $n \gtrsim 8$ [J. R. Hiskes and C. B. Tarter, Lawrence Radiation Laboratory Report UCRL-7088, Rev. I, 1964 (unpublished)] are candidates. From measurements by Kingdon et al. [J. Kingdon, M. Payne, and A. C. Riviere, UKAEA Culham Laboratory Report CLM-PR 11, 1968 (unpublished)] and Solov'ev et al. (E. S. Solov'ev, R. N. Il'in, V. A. Oparin, and N. V. Fedorenko, Zh. Eksp. Teor. Fiz. 53, 1933 (1967) [English Transl.: Soviet Phys. JETP 20, 1097 (1968)]] we conclude that less than 2% of $\rm H_2$ formed by electron capture by $\rm H_2^{+}$ in Mg at 40 keV is in hydrogen-like states with $n \geq 8$. We observe excited fractions on the order of 20% at this energy and attribute them to the $c^3\Pi_u$ state.
- 6. P. M. Stier and C. F. Barnett, Phys. Rev. 103, 896 (1956).
- 7. G. W. McClure, Phys. Rev. 134, A1226 (1964).

FIGURE LEGENDS

- Fig. 1. Attenuation of a beam of D₂ molecules, produced in a Mg neutralizer (thin target) by electron capture by 50-keV D₂⁺, in an H₂ target. The solid line connects the experimental data. Curve A is an extrapolation of the thick target asymptote; curve B is the difference between the data and curve A. •, experimental points; o, derived points.
- Fig. 2. Total-loss and ionization cross sections for collisions of energetic H_2 $\chi^1\Sigma_g^+$, σ_t and σ_i , and H_2 $c^3\Pi_u$, σ_t^* and σ_i^* , with H_2 gas. Present results: \blacktriangle , σ_t^{*} ; \bullet , σ_i^{*} ; Δ , σ_t ; σ_i^* . The results of McClure (Ref. 7) are indicated by \square . The lines have no significance other than to guide the reader's eye between data points. They should not be interpreted to portray any functional dependence. (Clearly σ_i^* $\leq \sigma_t^*$.)

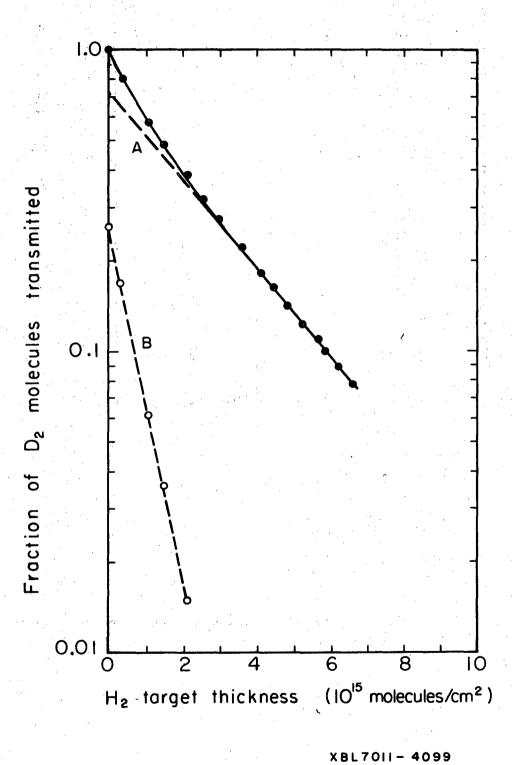
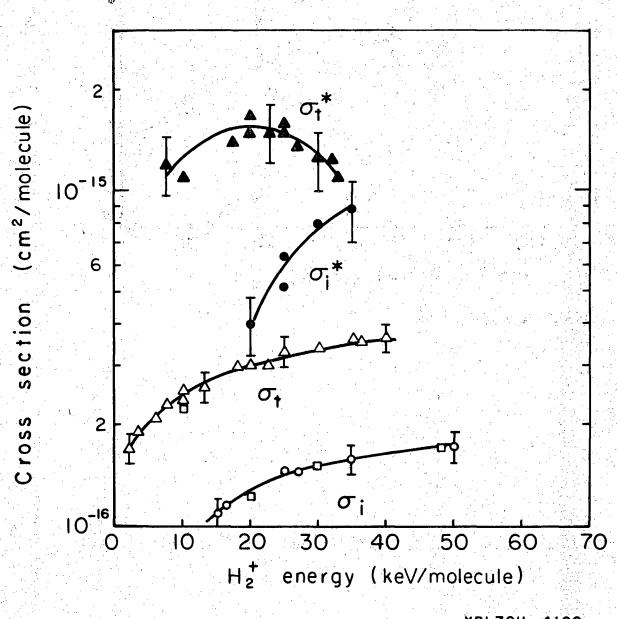


Fig. 1



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Fig. 2

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