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Measurement of the Total Cross Section for Charge Transfer into the Metastable State H(2s) for Proton Collisions with Atomic Hydrogen

James E. Bayfield
Gibbs Laboratory, Yale University, New Haven, Connecticut 06520
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The total cross section $\sigma_{2s}(H)$ for $H^+ + H(1s) \rightarrow H(2s) + H^+$ has been measured over the energy range 3–70 keV. Above 10 keV, absolute cross-section values are believed accurate to within ±35%. Only one maximum is found in the energy dependence of $\sigma_{2s}(H)$. The position of this maximum lies at an energy consistent with the Massey criterion. A comparison with theoretical cross sections suggests that intermediate-state coupling through the resonant charge-transfer channel $H^+ + H(1s) \rightarrow H(1s) + H^+$ is unimportant at low energies. The low-energy data is consistent with a molecular picture of the collision. Calculations utilizing Sturmian wave functions are not in agreement with the data. The Born approximation cross section lies significantly below experiment between 20 and 70 keV.

INTRODUCTION

The study of H^+ - H scattering at kinetic energies in the keV region is of special interest for three reasons. One-electron systems are most amenable to quantitative theoretical treatment. The rearrangement part of the scattering is separately observable. In addition, there is a good possibility that the coupling mechanisms responsible for specific collision-induced electronic transitions can be understood in some detail.

The three-or-more body problem does not permit exact solution. Consequently, any theoretical consideration of atomic collision phenomena involves semiempirical approximations which must be tested by experiment. Such approximations are most easily visualized for the three-body systems. For the H^+ - H system at energies above 10 eV or so, the relative motion of the protons is essentially classical. This fact further simplifies the theoretical problem.

For a keV proton beam incident on a target containing stationary hydrogen atoms, the character of the screened Coulomb interaction gives rise to the scattering of almost all incident protons through only small angles in the laboratory frame, accompanied by relatively little momentum transfer to target protons. Thus, for purposes of total cross-section measurements, incident and target protons are distinguishable. The distinguishability is essentially independent of the motion of the relatively light atomic electrons. This permits the study of charge or electron transfer without noticeable interference with the elastic-scattering channel. Such investigations ultimately are tests of the theory of rearrangement collisions, as the forces between the three particles are well known.

For most atomic-collision systems, actual calculations of observable quantities involve effective scattering potentials in addition to wave functions expanded in some appropriate complete set of functions spanning Hilbert space. For the special case of the one-electron problem, the adiabatic potentials can be accurately obtained. Also, wave-function expansions can be taken as a sum of product functions $\psi = \sum_i F_i \psi_i$, where the ψ_i are exactly known electronic wave functions clearly appropriate for the problem. The F_i are unknown functions for the relative motion of the protons that must be determined from Schrödinger's equation in a manner consistent with the boundary conditions for scattering states. The ψ_i are usually, but not always, taken as exact wave functions for either H or H_2^+. Uncertainties in the numerical predictions of theory should then arise only because the number of terms in the expansion must be truncated to some finite set for computational reasons.

The principal difficulty in choosing the properly truncated basis set is connected with the fact that an infinite number of scattering channels are energetically open. At 7 keV, experiment has shown that ionization of the target atom readily occurs; hence one concludes that excitation of the electron to all bound and many continuum states does take place to some extent. This situation can be treated formally using multichannel scattering theory, and creates no real computational difficulties unless a large number of channels are strongly coupled. If coupling is important, a judicious choice of the truncated set of basis functions is necessary. Thus, the coupling problem lies at the focus of much theoretical activity in the field of ion-atom scattering. Appropriate experiments on the one-electron systems are essential in order that further progress be made.

The comparison of existing experimental data with theoretical results is complicated by the

coupled multichannel character of the problem. Experimentally, the best-studied observable is the total charge-transfer cross section $\sigma_{10}(H)$, obtained as a function of incident proton energy.[1-4] This quantity is however a sum over the cross sections for charge transfer into each excited state. Charge transfer into the ground state is a resonant collision process in the case of the H^+-H(1s) system. Even so, the large number of excited states available may make their over all production non-negligible at the percent level, except for very low energies. Many scattering approximations adequately explain the $\sigma_{10}(H)$ data, considering experimental error and the uncertainties arising from excited-state production. Total ionization has also been experimentally studied,[5,6] but comparison with theory again involves an integration over final states, as the spectrum of final electron energies was not measured.

Up to the present, then, the experiments directly bearing on the coupling problem are three in number. Total direct and charge-transfer cross sections for Lyman alpha (Ly α) production have been measured between 0.6 and 30 keV[7]; the energy dependence of the total cross section $\sigma_{2s}(H)$ for charge transfer into the metastable state has been obtained above 43 keV,[8] and the fractional probability P_0 for charge transfer regardless of excitation has been measured for laboratory scattering angles between 0.2 and 6 deg and for incident energies between 0.13 and 150 keV.[9]

The importance of coupling to the $n=2$ states was suggested by the experimental results for P_0; the inclusion of $2p$ states in expansions employing hydrogen-atom basis wave functions (or the $2p\pi_u$ state for the H_2^+ basis set) was necessary but not sufficient to bring experiment and theory[10,11] into agreement. The importance of $2p$ states was confirmed in the Ly-α cross-section measurements, for the measured cross section was found to be 0.2 Å^2 at the relatively low energy of 0.6 keV. At 25 keV, the probability for Ly-α production in a charge-transfer collision is about 7%. Recent theoretical calculations of $\sigma 2p(H)$ qualitatively agree with these results.[10,12,13]

The results of calculations of the cross section $\sigma_{2s}(H)$ for charge transfer into the $2s$ state depend strongly on the truncated basis set used.[12-14] At energies above 40 keV, all theories predict a monotonically decreasing energy dependence of $\sigma_{2s}(H)$, which has been verified.[8] The objectives of the present experiment were twofold. First, the absolute magnitude of $\sigma_{2s}(H)$ was to be measured above 50 keV, where theoretical values span a factor of 8. Second, the energy dependdence and absolute magnitude of $\sigma_{2s}(H)$ were to be measured to as low an energy as possible, since theory appears quite sensitive to coupling between scattering channels below 7 keV. A report of preliminary findings has been published.[15]

APPARATUS

The experimental method used was basically that of earlier studies on charge transfer into the $2s$ state for protons incident on H_2 and Ar. The details of the apparatus are contained in the report of that work,[16] hereafter called Paper I. The principal change is the use of an atomic-hydrogen scattering target. A detailed report of the design and properties of the target has been made.[17] The cross section $\sigma_{2s}(H)$ was measured at each energy relative to the cross section $\sigma_{2s}(Ar)$ obtained in Paper I. The latter cross section is in agreement with the results of other investigations.[18,19]

A hydrogen-ion beam was obtained from a Thoneman radiofrequency-discharge ion source and was electrostatically accelerated to the desired energy. After mass analysis in a 60° magnetic spectrometer and collimation by two 0.127-cm diam apertures placed 70 cm apart, the beam was passed through the gas-scattering target. This incident pure proton beam had an intensity between 5×10^{-10} and 10^{-8} A, measured to within ±1%. The beam possessed an energy spread measured to be less than 100 eV, and the mean beam energy was known to within ±200 eV. Experiments confirmed the high collimation of the beam; the full width at half-maximum of the beam shape was 0.09°.

The scattering target was that gas residing within a differentially pumped cylindrical volume located inside a tungsten scattering cell. Atomic hydrogen was produced inside the cell by thermal dissociation at pressures of about 3×10^{-5} Torr and temperatures of about 2400°K. The cell was heated inside a vacuum furnace by the absorption of thermal radiation. The heater element of the furnace was driven by a source of rectified ac power. The 700-A output of the power supply was gated on for 68 msec and then off for 34 msec; the beam was gated on during the heater current off time so that the scattering occured in a region free of magnetic fields.

The fraction of H_2 molecules dissociated into atoms inside the hot cell was measured[17] using a modified version of the double charge-transfer technique introduced by Everhart and co-workers.[20] The dissociation fraction was 0.96±0.04. The nonhydrogen impurity level was believed to be below 0.2%.

Geometrical definition of the target region was improved through use of the target gas-flow bypass technique described in Paper I. Scattered atom signals were obtained first for the measured gas flow passing through the scattering cell, and second for the flow passing directly into the target vacuum-pumping chamber. Difference signals were obtained by subtracting the latter from the former. Such background-subtracted signals al-

most completely arise from scattering strictly inside the cell. A small contribution is due to gas in the cell differential-pumping channels. The weak atomic beam issuing from the cell yields a negligible contribution.

The methods used to detect scattered fast atoms were identical to those employed in Paper I. Metastable atoms were detected by Stark-effect quenching in a dc electric field and measurement of the resultant Ly-α radiation by a magnetic photomultiplier. In addition, fast atoms were detected without regard to their state of excitation using secondary electron emission from a metallic surface. Pulse counting was used in the metastable-atom detection channel; dc currents were measured in the neutral-atom detection channel.

Preliminary studies of scattered-atom angular distributions were studied as in Paper I. For atomic hydrogen in the target, the metastable beam was again found to be somewhat broader than the total neutral-atom beam. A typical beam scan is shown in Fig. 1. Such scans provided the information necessary to calculate the solid angle acceptance necessary for total cross-section measurements. Again, corrections of never more than 40% were made at the lowest energies, where the solid-angle acceptance could not be made quite large enough. Such corrections were based on the beam-scanning curves.

A search was made for quenching of metastable atoms inside the hot scattering cell. Such quenching might arise from collisions with thermionic electrons or thermally produced photons. These effects were found to be small through comparison of scattering data for cold and hot argon. In Fig. 2, the ratio $\sigma_{2s}(\mathrm{Ar})/\sigma_{10}(\mathrm{Ar})$ is plotted versus energy for the furnace both off and at full power. Within experimental error, no evidence of furnace quenching is seen. The data shown were taken in a more-or-less random manner over a period of months; the scatter in the points is mainly an indication of the stability of the detection systems used in the experiment.

All fast atom signals were shown to be linear with beam current and target gas flow to within

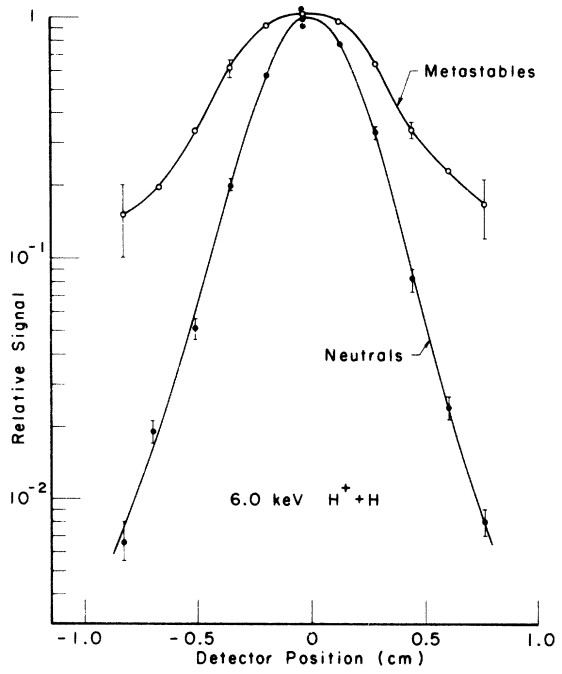

FIG. 1. Profiles of the scattered-atom beams. A long slit was placed in front of the atom detectors. Slit and detectors were simultaneously translated across the beam.

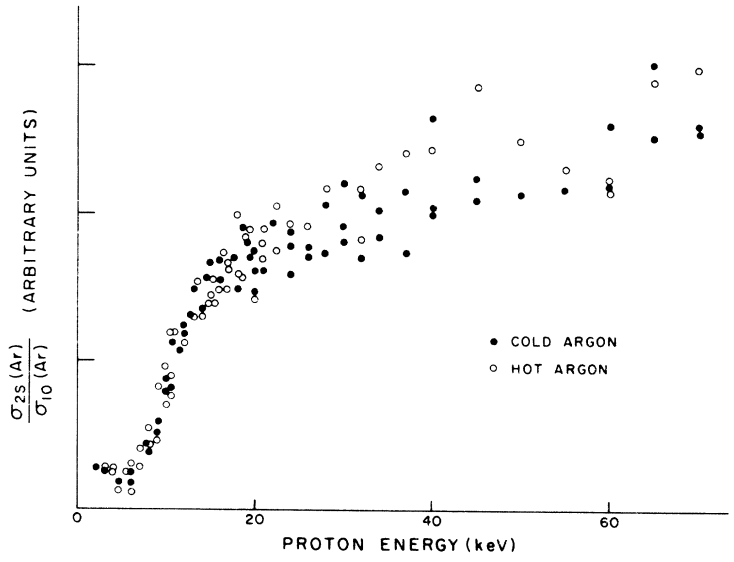

FIG. 2. Cross section ratio σ_{2s} (Ar)/σ_{10} (Ar). Dots denote data for the target at room temperature; circles are for data taken at 2400°K.

±2% for the ranges of values used in cross-section runs. This was established for both H and hot Ar target gases. By alternating target gases, the cross section $\sigma_{2s}(H)$ was measured relative to $\sigma_{2s}(Ar)$ for a given beam energy. The cross-section ratio $\sigma_{10}(H)/\sigma_{10}(Ar)$ was measured at the same time.

The background-subtracted metastable atom signals for hydrogen and argon in the heated scattering cell are, respectively,

$$F_*(H, H_2) = \eta^*[\sigma_{2s}(H)\Pi(H) + \sigma_{2s}(H_2)\Pi(H_2)],$$

$$F_*(Ar) = \eta^*\sigma_{2s}(Ar)\Pi(Ar),$$

where η^* is the metastable atom detection efficiency and the Π's are background-subtracted target thicknesses. We have $\Pi = nl$, where n is the mean target density and l is an effective target length of approximately 5 cm. Conservation of the flow dn/dt of molecules through the gas-handling system at temperature T_0 and target cell at temperature T yields the relations

$$\dot{n}(H_2) = \frac{1}{kT_0} Q(H_2) = \frac{1}{l} C_c(H_2)\Pi(H_2) + \frac{1}{2l} C_c(H)\Pi(H),$$

$$\dot{n}(Ar) = \frac{1}{kT_0} Q(Ar) = \frac{1}{l} C_c(Ar)\Pi(Ar),$$

where the C_c are hot-cell molecular flow conductances
$H/Ar = (40)^{1/2}$. The gas flows Q were measured as in Paper I. By definition of the measured dissociation fraction f,

$$n(H_2) = [2(1-f)/f]n(H) \equiv Kn(H).$$

The relations given imply that

$$S_* \equiv \frac{F_*(H, H_2)}{F_*(Ar)} \frac{Q(Ar)}{Q(H_2)} = \frac{\sigma_{2s}(H) + K\sigma_{2s}(H_2)}{(10)^{1/2}\sigma_{2s}(Ar)} \frac{1}{1 + (2)^{1/2}K},$$

which can be inverted to give the result

$$\sigma_{2s}(H) = (10)^{1/2} S_* \sigma_{2s}(Ar) + K[(20)^{1/2} S_* \sigma_{2s}(Ar) - \sigma_{2s}(H_2)].$$

Since S_* and K are measured quantities, $\sigma_{2s}(H)$ can be obtained from known values of $\sigma_{2s}(Ar)$ and $\sigma_{2s}(H_2)$. A similar expression relates $\sigma_{10}(H)$ to $\sigma_{10}(Ar)$ and $\sigma_{10}(H_2)$. The bracketed term in the above expression contains two corrections that partially cancel each other. They arise because incomplete hydrogen dissociation ($K \neq 0$) results in a decrease in $\Pi(H)$, giving the first term, and its replacement by $\Pi(H_2)$, giving the second term. The value of $\sigma_{2s}(H)$ depends only slightly on $\sigma_{2s}(H_2)$ because $K = 0.08$ and $\sigma_{2s}(H_2)$ is never much larger than $\sigma_{2s}(H)$. A similar situation holds for the dependence of $\sigma_{10}(H)$ on $\sigma_{10}(H_2)$. The values of $\sigma_{10}(Ar)$, $\sigma_{10}(H_2)$, $\sigma_{2s}(Ar)$, and $\sigma_{2s}(H_2)$ used to compute $\sigma_{10}(H)$ and $\sigma_{2s}(H)$ are given in Paper I.

The values of $\sigma_{10}(H)$ presently obtained are compared in Fig. 3 with the data of previous absolute measurements.[1,2] The stated uncertainty of the data of McClure[2] is ±5%. The present data are believed accurate to ±10% above 10 keV and to ±15% below 10 keV. The largest contribution to these error estimates is the uncertainty in $\sigma_{10}(Ar)$ discussed in Paper I. The values obtained for $\sigma_{10}(H)$ are in good agreement with earlier work. This study of total charge transfer can be considered as an additional search for possible errors inherent in the experimental method used to measure $\sigma_{2s}(H)$. No evidence of systematic effects is found.

RESULTS

The data obtained in the present experiment are shown in Fig. 4. The values of $\sigma_{2s}(Ar)$ used to

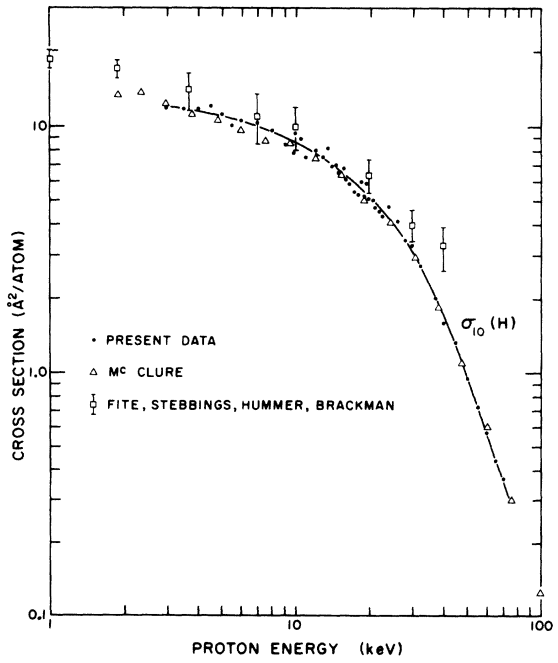

FIG. 3. Total charge-transfer cross section $\sigma_{10}(H)$. The uncertainty in the present data is about ± 10% above 10 keV, and ±15% below 10 keV.

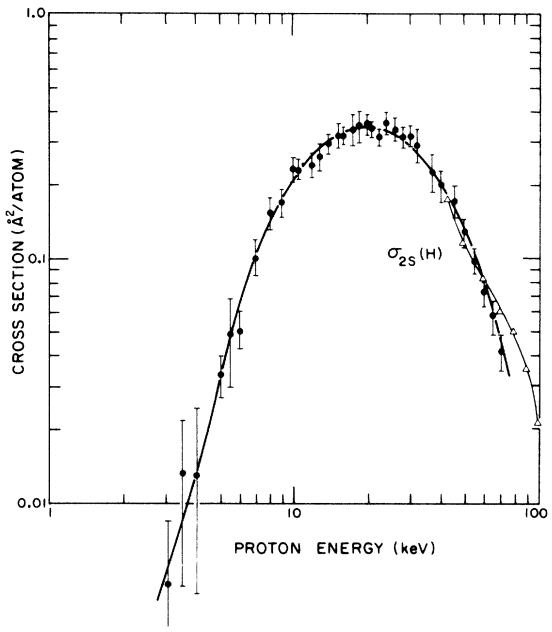

FIG. 4. Cross section σ_{2s}(H). The error bars represent the over-all uncertainty in the energy dependence. The absolute magnitude of the cross section has an additional uncertainty of about ±25%. Diamonds denote the relative data of Ryding, Wittkower and, Gilbody normalized to the present data at 43 keV.

calculate σ_{2s}(H) were obtained from a smooth curve drawn through the data points of Fig. 7 in Paper I. The only uncertainty not included in the error bars is the uncertainty in the cross-section scale associated with the value of σ_{2s}(Ar) at 20 keV. This quantity is believed known to within ±25%. Thus the absolute cross section at most energies is known to within about ±35%. The solid line is hand drawn through the data points.

At energies above 40 keV the energy dependence obtained by Ryding, Wittkower, and Gilbody[8] can be compared with the present data. Their results for the relative cross section are shown in Fig. 4. Normalization of their curve is to the value presently obtained at 43 keV. The uncertainty in the relative values of Ryding *et al.* is about ±25%. Thus, the two experiments are in agreement.

At low energies the fractional uncertainty in the measured value of σ_{2s}(H) becomes relatively large. This is primarily because the magnitude of the cross section becomes quite small, resulting in poor signal-to-noise ratios. Also, at 3 keV a possible error of about ±20% arises from uncertainty in the correction made for incomplete metastable atom passage through the detector aperture. It should be pointed out that if σ_{2s}(H) were about 0.1 Å² at 3 keV instead of an order of magnitude smaller, it could have been measured to approximately the accuracy achieved for σ_{2s}(Ar) in Paper I. The data below 7 keV have been reproduced four times in runs occurring months apart.

DISCUSSION

The Massey criterion predicts that, in the absence of level crossings, an atomic collision cross section will be maximal when the collision time is about equal to the time for transition between the relevant states. Thus, $a/v \sim h/\Delta E$, where here $\Delta E = 10.2$ eV is the energy defect for the collision and a is found empirically to be about 7 Å.[21] The velocity v computed by this formula corresponds to a proton energy of 17 keV. The experimental maximum for σ_{2s}(H) is between 20 and 25 keV. We conclude that the observed scattering is due to the fundamental collision resonance for the process, occurring in the expected energy region.

Early calculations of σ_{2s}(H) employed the first Born approximation.[22] This theory includes only initial and final atomic states for the electron and therefore ignores the coupling problem. The relative motion of the protons is treated quantum mechanically as that of a free particle. It is commonly believed on physical grounds that the Born approximation should be valid at sufficiently high energies, ($a/v \ll h/\Delta E$). Expressing this mathematically, the Born approximation is the first term of an expansion representing the collision as a perturbation of an incident plane wave. However, in the present case of a rearrangement collision, the convergence of the expansion is in question.[23] A comparison of the Born result with the present experiment can be made (see Fig. 5). Below 35 keV the theory is in qualitative disagreement with the data. At high energies the experimental curve appears to be approaching the Born curve at 70 keV. On the other hand, normalization of the relative data of Ryding *et al.*[8] to the present value at 43 keV gives an experimental value for σ_{2s}(H) at 120 keV of about twice the Born value. Data at still higher energies is needed for a test of the Born approximation in its energy region of expected validity.

Several points computed[24] with inclusion of the continuum part of the second term in the Born series are also shown in Fig. 5. This procedure increases the disparity between theory and experiment, as low energy values increase and high energy values decrease relative to the first Born results.

Calculations based upon the impact-parameter method (IPM)[25] appear capable of better agreement with experiment. This theoretical approach is a generalization of Dirac's method of variation of constants; an assumed classical motion of the

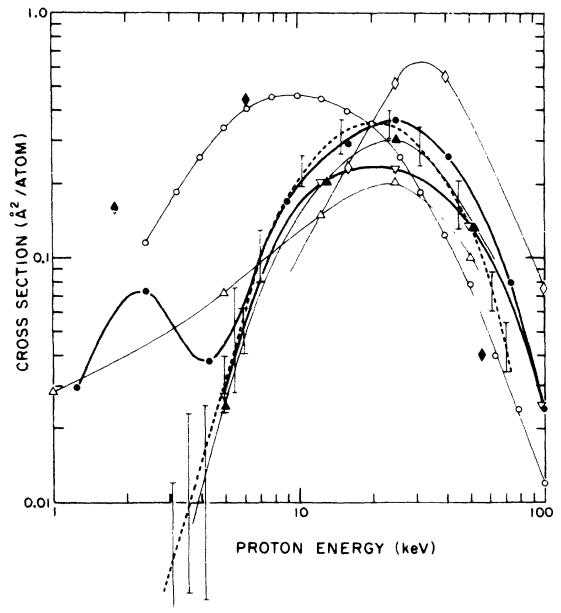

FIG. 5. Comparison of theoretical predictions with the present data. ---- The data, with total uncertainty indicated by error bars. The approximations made in the theoretical curves are: O Born; ♦ second Born; ▽ $1sA/2sB$; ▲ $1sA/2sA/2sB$; △ $1sA/1sB/2sB$; ● four-state hydrogenic close coupling; ⟠ four-state Sturmian close coupling.

protons is used to replace the time dependence of the problem with a dependence on internuclear separation. The relative proton motion is usually but not necessarily taken to be rectilinear and constant in velocity.

Up to the present, IPM calculations of $\sigma_{2s}(H)$ have employed expansions of the system wave function based upon atomic wave functions centered around both the target proton A and the incident proton B. The simplest treatment is to ignore couplings to intermediate states and include only the initial and final electronic states ($1sA$ and $2sB$) in the expansion. The results obtained[12] in this approximation are labeled $1sA/2sB$ in Fig. 5. The energy where the maximum in the cross section occurs is in close agreement with experiment, as is the behavior at low energies. This approximation lies significantly below the data for energies between 10 and 25 keV and noticeably above the data for energies greater than 60 keV.

The importance of the coupling problem can now be seen by comparing the data with IPM calculations retaining additional states in the eigenfunction expansion. The inclusion of the $2sA$ state[12] raises the cross section between 10–25 keV to values in agreement with experiment, while not disturbing the low-energy behavior. This $1sA/2sA/2sB$ result produces the best agreement with experiment to date. On the other hand, inclusion of the $1sB$ state[12] (see the curve labeled $1sA/1sB/2sB$ in Fig. 5) moves theory away from experiment at all energies. Close-coupling calculations[13] using $1s$, $2s$, $2p_0$, and $2p_{\pm1}$ states about each proton are in good agreement with the data, except for the prediction of an unobserved secondary maximum near 2 keV.

The use of Sturmian wave functions[14] instead of the usual atomic eigenfunctions in four-state ($1s$, $2s$, $2p_0$, $2p_{\pm1}$) close-coupling calculations results in values of $\sigma_{2s}(H)$ in marked disagreement with the present data. Adding the $3s$ and $4s$ Sturmian states[14] improves the theory at 40 keV, but not very much at 25 keV.

Understanding of the coupling-sensitive Ly-α cross section and large-angle scattering experiments mentioned in the introduction is based upon wave mechanical treatments[10, 11] employing molecular basis functions. At present, it appears[26] that it is not so much the classical character of the IPM that should be questioned as it is the use of atomic eigenfunctions in describing phenomena that can occur in the H_2^+ system at small internuclear separations. That the low-energy behavior of $\sigma_{2s}(H)$ may be a case in point can be seen by referring to the energy-level diagram of the Born-Oppenheimer electronic states of H_2^+ (Fig. 6).[27]

One should note that the H_2^+ system is special in that the nuclei are identical; this leads to states of definite parity. Transitions between *molecular* energy levels must conserve parity.

The initial state for the present scattering problem is a linear combination of the $1s\sigma_u$ and $2p\sigma_u$ molecular states, and thus is a molecular state of mixed parity. Production of $H(2s)$ must involve transitions between the two initial molecular states and some of the six higher-lying excited states. At sufficiently low energies, transitions from the $1s\sigma_g$ state are relatively unlikely[28] because the separations between this state and the excited states of even parity are too large. Thus, transitions from the $2p\sigma_u$ state are probably of major importance. Such transitions going to the $2p\pi_u$ state are induced by rotation of the internuclear axis, and are in fact responsible for the observed large low-energy values of $\sigma_{Ly\text{-}\alpha}$ and some of the damping of P_0. However, population of the $2p\pi_u$ state cannot produce $H(2s)$ unless further transitions with other levels occur. The energy splitting H_{12} at the $2p\pi_u$ - $4f\sigma_u$ crossing is probably determined by rotational coupling terms in the Hamiltonian. Because of the large value of the internuclear separation at this crossing, H_{12} is expected to be small, and the passage of the system through the crossing nearly diabatic. Thus a population of the molecular levels leading asymptotically to the atomic $2s$ state requires either transitions between widely separated levels or double transitions involving rotationally coupled

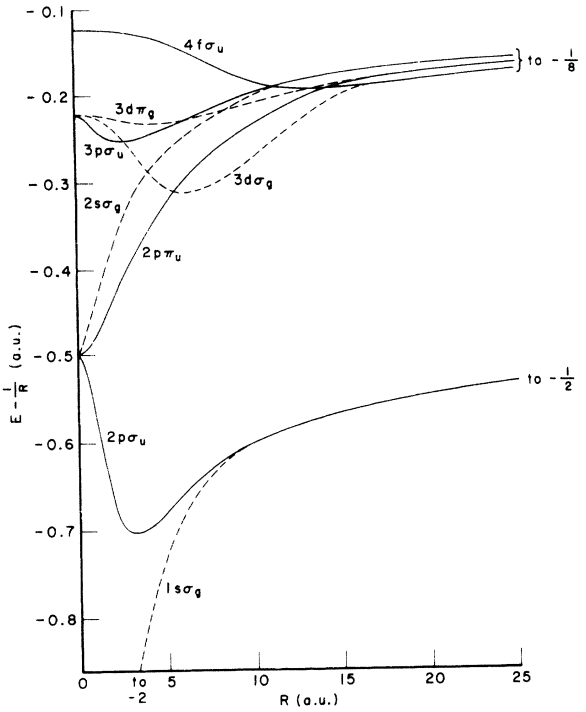

FIG. 6. Lower electronic energies of the bound states of H_2^+. The internucear repulsive Coulomb potential is not included. The different states are labeled by their united-atom designations. Atomic units are used.

levels, both relatively unlikely occurrences. One therefore expects $\sigma_{2S}(H)$ to be small at low energies, in agreement with the data.

The 2-keV maximum in $\sigma_{2S}(H)$ predicted by the four-state hydrogenic close-coupling theory appears similar to the secondary maxima experimentally observed in σ_{2S} for proton collisions with the inert gases and with H_2 (see Paper I). In contrast to the H_2^+ case, in these other systems parity is not a good quantum number. Transitions between the molecular states $2p\sigma$ and $2s\sigma$ may therefore occur as the system approaches small internuclear distances. Thus $\sigma_{2S}(H)$ can be expected to exhibit a low-energy behavior quite different from that of $\sigma_{2S}(H_2)$ or $\sigma_{2S}(Ar)$.

The possible equivalence of an H_2 molecule to two H atoms in keV atomic collision phenomena has been of interest for some time. Sizable differences are expected theoretically[29] and are observed[2,3] for the case of total charge transfer for incident protons. The present data for $\sigma_{2S}(H)$ can be compared with the data for $\sigma_{2S}(H_2)$ obtained in Paper I (see Fig. 7). The molecular effects present in H_2 are seen to be unimportant only for energies of 70 keV or higher. The present data agree well with those of Ryding *et al.*[8] over the energy range common to both experiments.

The discussion presented is based at several points upon the assumption that one can ignore cascade contributions to $\sigma_{2S}(H)$ arising from charge transfer to high excited states and subsequent decay to the 2s state. Little concrete information is available on this point, as theoretical estimates cannot be considered quantitative and experimental studies for states with $n > 2$ have not been made. The problem of cascade effects is in fact an important source of uncertainty inherent in the comparison of experimental data with theoretical predictions at the higher energies. Calculations based upon Born approximation cross sections[21] have been made; the cascade contribution to $\sigma_{2S}(H)$ so obtained is about 6% at 20 keV.

The present measurements can be used in conjunction with the earlier measurements for charge transfer into the 2p state[7] to estimate the cascade contributions to the total charge-transfer cross section $\sigma_{10}(H)$. A lower bound is found to be 16% at 30 keV and 2% at 3 keV.

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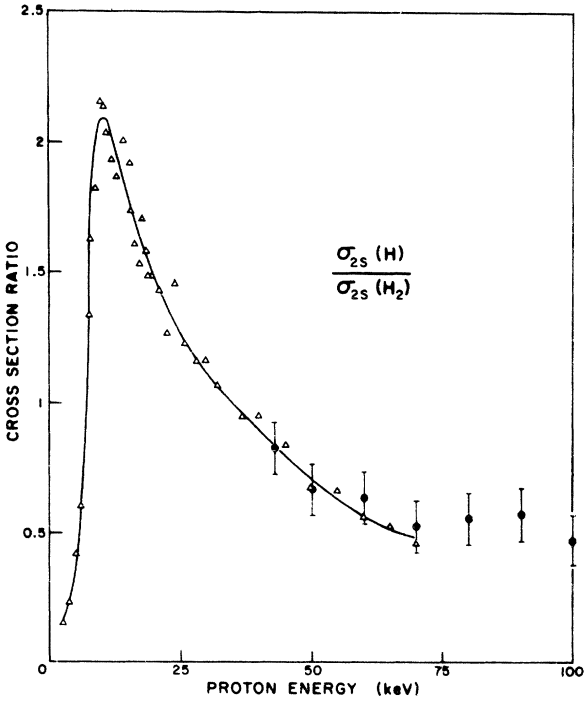

FIG. 7. Cross-section ratio $\sigma_{2S}(H)/\sigma_{2S}(H_2)$. If a hydrogen molecule were equivalent to two atoms, the value would be $\frac{1}{2}$. Δ Present data; Φ Ryding, Wittkower, and Gilbody.

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