Collisional Quenching of Metastable Hydrogen*†

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Measurements of the collisional quenching of the 2*S* state of atomic hydrogen by helium, neon, argon, and krypton have been made. The energy range was in the thermal region ($v \sim 10^6$ cm/sec). The cross sections were determined by measuring the attenuation of a beam of H(2*S*) atoms in passing through targets of the various gases. Time-of-flight techniques were used to measure the velocity dependences of the cross sections. For an incident H(2*S*) velocity $\approx 1\times 10^6$ cm/sec, the measured cross sections are σ(helium) = 81 ± 12 Å^2/atom, σ(neon) = 80 ± 12 Å^2/atom, σ(argon) = 91 ± 14 Å^2/atom, and σ(krypton) = 108 ± 16 Å^2/atom. The results for helium are 10–15% higher than theoretical results obtained by Slocomb, Miller, and Schaefer and 20–25% higher than a calculation by Byron and Gersten using a pseudopotential method.

I. INTRODUCTION

The study of the quenching (depopulation) of hydrogen in the metastable 2*S* state in collisions with simple target atoms is of considerable importance in that the measured cross sections can be compared with results calculated from first principles. If the relative energy of the collision partners is sufficiently low, these collisions should truly be simple inelastic collisions, simple in the sense that only a few states need be considered.

Byron and Gersten[1] and Slocomb, Miller, and Schaefer[2] have reported calculations for the quenching cross sections of metastable hydrogen in collisions with helium targets in the energy range roughly below 250 eV. At these energies they considered only the 2*S*-2*P* transitions. The Byron–Gersten calculation formulates the problem in a time-dependent framework. Two approximations to the potential are used: a pseudopotential approach, and a simple summation of the electrostatic interactions, referred to as the perturbation method. In the Slocomb–Miller–Schaefer paper, the cross section is derived in a partial wave approach with appropriate Born–Oppenheimer interaction potentials.

Experimental studies in the thermal region should provide a rigorous check on these calculations since the quenching in this region should arise only from these 2*S*-2*P* transitions. We report here the detailed results of measurements of the quenching of H(2*S*) in collisions with helium, neon, argon, and krypton targets in the thermal energy region. Preliminary results for quenching H(2*S*) by helium were reported in an earlier paper.[3] Results for low-energy quenching of H(2*S*) by helium have also been reported by Comes and Wenning.[4] At higher energies measurements have been reported by Byron *et al.*,[5] Spiess *et al.*,[6] Gilbody *et al.*,[7] and Dose *et al.*[8]

In Sec. II the experimental apparatus and procedures used in measuring these cross sections are described. The results of the experiment are discussed in Sec. III.

II. EXPERIMENTAL

A. Apparatus

The experiment was conducted in a cylindrical brass vacuum system which was divided into two differentially pumped regions, as shown schematically in Fig. 1. Both chambers were evacuated with 4-in. oil diffusion pumps baffled with liquid-nitrogen cold traps. The source chamber was maintained typically at 5×10^{-5} torr while the scattering/detection region was kept typically at 7×10^{-7} torr.

Atomic hydrogen was produced by thermal dissociation of molecular hydrogen in a tungsten oven. The oven, a 0.254-cm-diameter tungsten tube with a 0.051×0.013-cm slit, was heated to ~2800 °K. The oven temperature was monitored with an optical pyrometer. The oven pressure was typically 5 torr. The gas effusing through the slit was typically 70–80% dissociated as expected at this temperature and pressure. The diffuse beam of ground-state atomic and molecular hydrogen was then collimated by a slit (typical slit width = 0.1 cm) which separated the source chamber from the excitation region.

The collimated atomic- and molecular-hydrogen beam was cross-fired with a pulsed electron beam of ~13 eV energy producing both metastable atoms and molecules. The electron gun used was a simple diode mounted within a collimating electromagnet. Typical dc emission current density was 2 mA/cm^2 with the collimating magnet on. The anode was heated to 150 °C to prevent condensation of pump oil.

The beam then passed into the scattering region

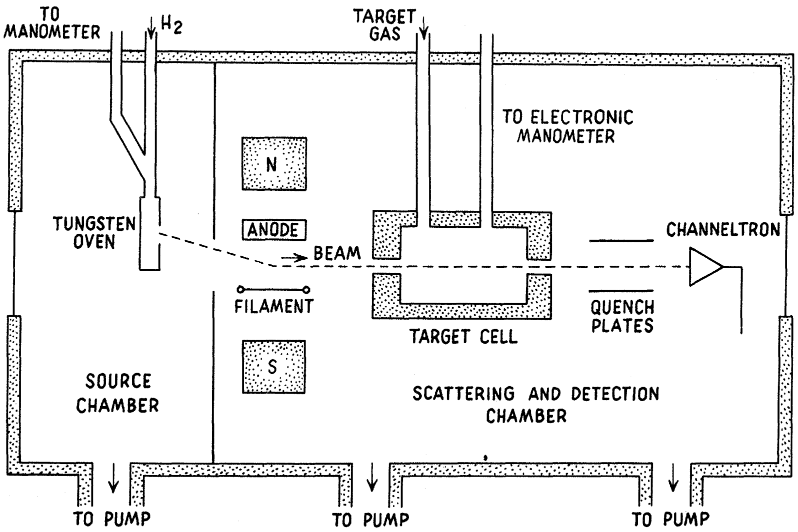

FIG. 1. Atomic-beam-machine schematic.

through another collimating slit (0.076-cm-diam hole) covered with a grounded mesh to trap charged particles.

The scattering cell was a simple cylindrical chamber 2.54 cm in diameter and 7.23 cm in length. The entrance channel was cylindrical (0.126 cm diam, 0.890 cm long) and the exit consisted of interchangeable cylindrical (0.254 cm diam, 0.890 cm long) and rectangular ($1.52\times 0.101 \times 0.890$ cm) slits. The rectangular slit was used to determine the effects of elastic scattering on the H(2S) beam. Flow of the target gas from a 1-liter reservoir was regulated with a commercial variable leak. Target pressures were typically held constant to $\pm 3\%$.

Initial measurements of helium pressures in the target cell were made with an ionization gauge calibrated against a mercury manometer which was trapped with liquid nitrogen. However, most of the target pressure readings were conducted with a Datametrics model No. 1014 electronic manometer together with a Barocel model No. 521 pressure sensor. The advantage of such a manometer is that the calibration is independent of the type of gas being studied.

The detector was a Bendix Channeltron electron multiplier. Metastable atoms and molecules and ultraviolet photons created during the excitation process eject electrons with a probable low efficiency[9] upon striking the detector surface. The Channeltron, mounted in an aluminum box to reduce noise, was approximately 0.85 cm in diameter. It was mounted on a movable arm which permitted rotation in a horizontal circle centered at the target chamber. This, combined with slits mounted on the detector box, permitted measurements of the angular distribution of the beam.

Following the target cell was a quench region (two parallel copper plates) in which a dc field (150 V/cm) could be applied to produce quenching of the H(2*S*) beam by Stark mixing of the 2*S* and 2*P* states with subsequent decay to the 1*S* state. Using the quench technique to measure the so-called "flop," the difference in the metastable beam intensities with the applied quenching field off and on, the unwanted background arising from metastable H_2 and ultraviolet (1216-Å) photons was measured and subtracted. The typical "flop" counting rate was on the order of 2000/sec.

The velocity distribution of the metastable atom beam results in a spread in the times of flight between the electron gun and detector, a distance of 17.4 cm. This distribution is non-Maxwellian due to recoil effects during the excitation process.[10,11] However, the details of the nature of this distribution were not important in this experiment since individual velocities were measured. Conventional time-to-pulse-height conversion techniques were used to determine the H(2*S*) velocities. The entire system was synchronized to a recurrent pulse generator. The electron gun was pulsed with a 5-μsec-wide voltage pulse at a 12-kHz repetition rate. Signal pulses from the electron multiplier were amplified, shaped, and used to trigger a linear gate operating in the coincidence mode. These signal pulses were also fed into a signal averager to monitor the beam-counting rate over all velocities. The gate input was a linear voltage ramp triggered simultaneously with the electron gun pulse. The gate output was a collection of pulses with voltage heights directly proportional to the time between metastable excitation and detection. These pulses were introduced into a Nuclear Data series 2200 multichannel analyzer operating in the pulse-height-analysis mode, where they were stored as a function of pulse height.

The pulse height is a function of the time of flight. In this manner, data were collected for given periods (typically 200 sec) and time-of-flight spectra were developed. Data stored in the analyzer were recorded on paper tape and analyzed on a PDP-10 computer.

B. Procedure

In passing through a target of stationary particles an incident monoenergetic beam of particles of flux I_0 and velocity v will be attenuated according to the familiar relation

$$I = I_0 e^{-nl\sigma_T(v)}, \tag{1}$$

where I is the emerging flux, n is the target density, l is the target length, and $\sigma_T(v)$ is defined as the velocity-dependent total scattering cross section. In general, however, $\sigma_T(v)$ describes more than one interaction. For the collisions of interest here, the total cross section can be written as a sum of the quenching cross section, i.e., the cross section for transitions out of the 2S state, and the elastic scattering cross section:

$$\sigma_T = \sigma_Q + \sigma_E. \tag{2}$$

The attenuation of the metastable-hydrogen beam was measured as a function of target gas density. Thus any contribution to a net decrease in metastable signal would be related to the total scattering cross section by (1). To determine the quenching cross section it was then necessary to determine the magnitude of the elastic scattering cross section.

To determine the velocity dependence of the measured cross sections, attenuation measurements were taken with the previously discussed time-of-flight techniques. In these measurements, the cylindrical exit channel (diameter of 0.254 cm) was installed in the gas cell; the detector slit was fully opened. Throughout these measurements, drifts in oven temperature, oven pressure, electron gun bombardment current, and collimating magnet field strength limited beam stability to ≈ 7%. Similarly, the pressure in the gas cell typically drifted 3–5% of the measured pressure. Therefore a long (5-μsec) voltage pulse on the electron gun was chosen to ensure a high counting rate and thus a reduction in the length of the counting periods. Beam flop was measured by storing Channeltron counts in the analyzer for 200 sec (quench off) and subtracting unwanted background counts (quench on) for equal periods.

At the beginning of each run, the channels of the multichannel analyzer were calibrated against time. Starting at the center of the electron gun voltage pulse, shaped pulses identical to shaped detector pulses, were fed into the linear gate at intervals of 5 μsec. The resulting gate output then consisted of equally spaced sections of the voltage ramp which, when fed into the analyzer, were recorded in channels which then represented the duration of the electron gun pulse and the corresponding calibrated intervals that followed.

I and I_0 were measured for six to ten densities for each target gas. To minimize the effects of beam drift the initial beam strength I_0 was measured before and after each beam measurement at a particular pressure. The average of these readings was taken as I_0.

To determine the effects of elastic scattering on the beam, a series of angular distribution measurements were made. Comparisons were made of the angular spread of the beam with the 0.254-cm-radius cylindrical scattering cell exit slit to the spread with the rectangular cell exit slit. In every case, the detector slit was closed to 0.125 cm. For each target gas, the angular distribution of the beam was recorded with the target cell evacuated and with a cell pressure approximately equal to a typical pressure used in the attenuation measurements. Counting with the signal averager over-all velocities, the flop was measured as a function of detector angle. The distributions were measured with respect to the center of the scattering cell, as the detector angles were calibrated through the rotation of the detector arm, pivoted at the cell center. For each geometry, a series of flop measurements were taken at angular intervals of 3.2 mrad and centered at the peak of the beam. For helium, extremely detailed angular distributions were measured. In this case, data were taken at angular intervals of 1.6 mrad. The angular spread was measured to about ± 3° from the center of the beam.

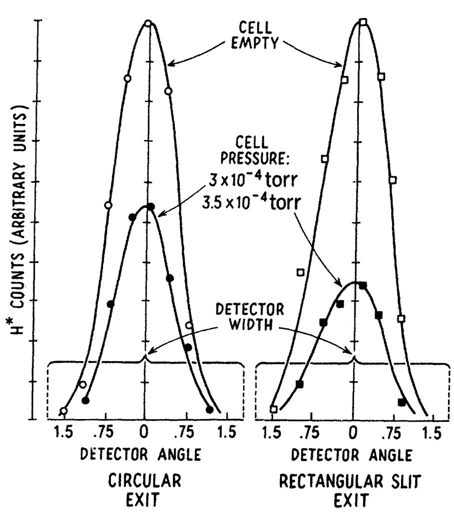

FIG. 2. Beam shape with cell empty and full using a helium target gas. The angular scale is in degrees.

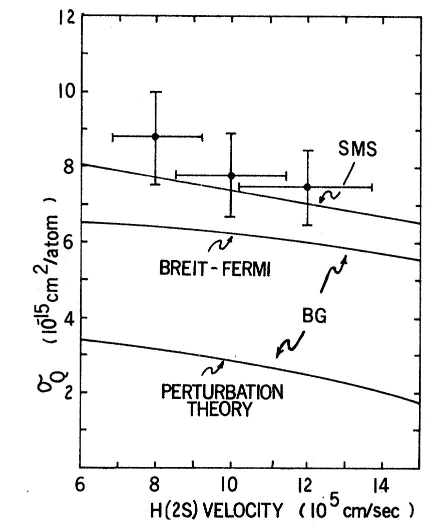

FIG. 3. Theoretical quenching cross-section curves (Byron–Gersten and Slocomb–Miller–Schaefer) with representative experimental points; the target gas is helium.

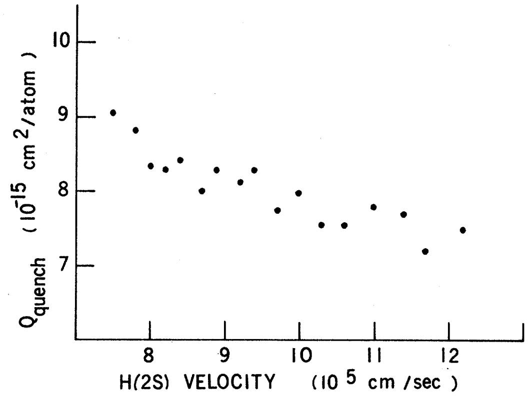

FIG. 5. Experimental cross section for quenching H(2S) by neon.

III. RESULTS

Cross sections were determined from the data in a channel-by-channel analysis of the data stored in the multichannel analyzer. Each channel corresponded to a particular value of H(2S) velocity. The values of I and I_0, for each scattering cell pressure and each channel (velocity), were least-squares fitted with the general equation

$$\ln(I/I_0) = -nl\sigma(v) + \text{const} . \qquad (3)$$

The slopes of these curves yielded the desired cross sections for each velocity analyzed. The constant term above represents the uncertainty in pressure measurements. Ideally, this constant would be zero but, in fact, varied from zero for the various data analyzed. In general, channels corresponding to intensities equal to or less than the full width at half-maximum of the H(2S) time-of-flight curves were not included in the cross-section analysis as the total number of counts stored in these channels were too low to ensure good statistics. Thus, each run typically yielded cross sections for about 15 channels (velocities).

A total of 17 runs were undertaken to observe elastic scattering effects on the beam. Four measurements were made of the beam shape with the scattering cell empty and the cylindrical exit channel installed; five profiles were studied with the cell empty and rectangular exit geometry employed. Similarly, measurements were made for each of the four target gases (at pressures approximately 3×10^{-4} torr) for both cylindrical and rectangular exit slit geometries. Pressure measurements for the slit geometry were not corrected for gas flow through the slit and were therefore only approxi-

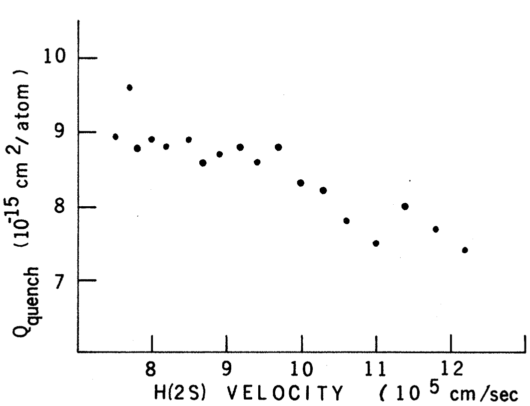

FIG. 4. Experimental cross section for quenching H(2S) by helium.

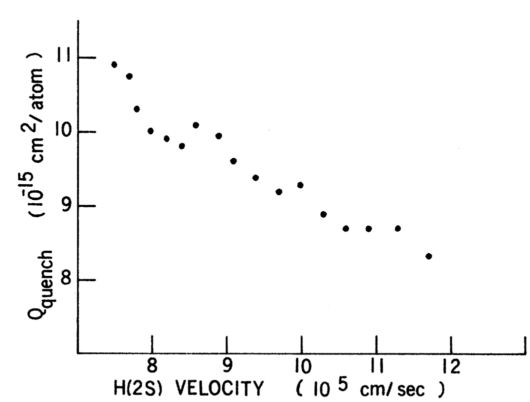

FIG. 6. Experimental cross section for quenching H(2S) by argon.

mate measurements. However, comparison of rectangular and cylindrical exit geometries for all gases showed no limitation in beam spread by the use of the cylindrical exit channel. It also showed that the attenuated beam was confined to the region intercepted by the detector. Therefore there was no measurable attenuation due to H(2S) atoms elastically scattered out of the detected beam. These results are indicated in Fig. 2. Since there were no measurable elastic scattering effects at the energies of this experiment, it was concluded that H(2S) atoms most likely are quenched before they are elastically scattered through angles greater than those subtended by our detector. This is in agreement with a simple semiclassical calculation.

Representative experimental data for quenching of H(2S) by helium are compared to the theoretical calculations in Fig. 3. The indicated experimental error in velocity is due to an uncertainty in the times of flight because of the finite pulse width on the electron gun and the finite length of the electron gun anode. The major contributions to errors in the cross sections are errors in pressure measurements, uncertainties in both I and I_0, and statistical errors in determining the weighted means of the data. The corrections for effective length of the scattering region and for the velocity distributions of the target gases introduced additional errors of only $\pm 2\%$.[12]

The weighted means of the measured quenching cross section for a helium target are 10–15% higher than the cross sections calculated by Slocomb, Miller, and Schaefer and are about 20–25% higher than the cross sections calculated by Byron and Gersten using their pseudopotential, or Breit-Fermi, approximation.

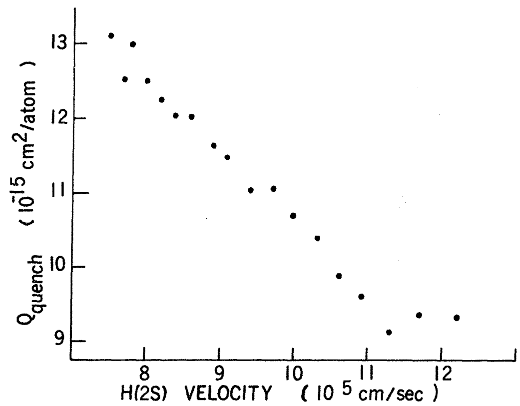

FIG. 7. Experimental cross section for quenching H(2S) by krypton.

Figures 4–7 show the cross sections for each of the four target gases used in the experiment. At this time, no detailed theoretical calculations are available for any target gases other than helium.

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