J. Phys. B: At. Mol. Opt. Phys. **29** (1996) 1219–1224. Printed in the UK

Formation of H(2s) atoms by excitation in 10–100 keV H^+–H collisions

D P Higgins, J Geddes and H B Gilbody

Department of Pure and Applied Physics, The Queen's University of Belfast, Belfast, UK

Received 16 November 1995

Abstract. Cross sections for 2s excitation of H atoms in 10–100 keV H^+–H collisions have been determined using a modulated crossed-beam technique. The measurements have been based on observations of the Lyman alpha radiation emitted during electric-field-induced decay of the metastable H(2s) collision products. The results extend the range of the 5–26 keV cross sections measured by Morgan and co-workers to intermediate energies where theoretical predictions based on close-coupling methods are known to be strongly dependent on the choice of the expansion basis. The present cross sections pass through a broad maximum at about 40 keV. Over the range 5–100 keV the available experimental data exhibit an undulatory structure similar to that predicted by some close-coupling calculations but good quantitative agreement is very limited. Close-coupling calculations which employ large basis sets and include a large number of projectile states at the expense of target states are shown to agree less satisfactorily with experiment than those which include only the dominant 1s capture projectile channel.

1. Introduction

A detailed understanding of collisions between protons and hydrogen atoms continues to be of considerable fundamental interest. The various inelastic collision processes are also relevant to the heating, modelling and diagnostics of controlled thermonuclear fusion plasmas and to the physics of the Earth's high atmosphere. In this work we consider the simple excitation process

$$H^+ + H \rightarrow H^+ + H(2s) \tag{1}$$

which has been the subject of numerous theoretical studies over a wide energy range. Accurate calculations are difficult, particularly at low and intermediate velocities where the theoretical models must describe the strong coupling between the excitation, electron capture and ionization collision channels (cf Reinhold *et al* 1990). Reliable experimental data can provide important checks on the range of validity of such calculations. Experimental studies of (1) have so far been very limited and confined to energies below 26 keV although measurements of cross sections for the corresponding 2p excitation process now extend to 800 keV (Detleffsen *et al* 1994).

In our previous work in this laboratory (Morgan *et al* 1973) we employed a modulated crossed-beam technique to obtain cross sections for (1) in the range 5–26 keV. Subsequently, Chong and Fite (1977), using a similar experimental approach, obtained data in the range 6–25 keV. In the present work, we have used a modulated crossed-beam technique in conjunction with a recently developed microwave discharge high-intensity source of H

0953-4075/96/061219+06$19.50

atoms (McCullough *et al* 1993) to obtain cross sections for (1) in the range 10–100 keV. Our measurements have been based on observations of the Lyman alpha radiation emitted during electric-field-induced decay of the metastable 2s atoms formed in the excitation process. The cross sections obtained complement the low-energy data of Morgan *et al* (1973) and extend to intermediate velocities where the results of calculations based on close-coupling methods are known to be strongly dependent on the expansion basis (Slim and Ermolaev 1994).

2. Experimental approach

2.1. General description

The apparatus and measuring procedure were similar to that used in our recent studies of $H(n = 2)$ excitation in He^{2+}–H collisions (Hughes *et al* 1994) so that only a summary of the essential features need be given here.

A momentum analysed and well collimated proton beam of selected energy within the range 10–100 keV was arranged to intersect at 90° a thermal energy beam of highly dissociated hydrogen from a microwave discharge source (McCullough *et al* 1993). Although atom beam densities greater than 10^{13} atoms/cm^3 at the source exit with dissociation fractions of about 0.9 were normally possible, as in our previous work, it was necessary to insert a Teflon baffle in the exit canal to reduce the amount of background Lyman alpha radiation emitted from the source; this reduced the dissociation fraction to about 0.75. The dissociation fraction was determined by using a quadrupole mass spectrometer to sample the hydrogen beam (Hughes *et al* 1994).

H(2s) atoms formed in the crossed-beam region were detected at a point 1.5 cm beyond the actual crossing region where an electric field of 15 V cm^{-1} was applied along the atom beam path by means of a pair of high transparency grids (see figure 2 of the paper by Hughes *et al* 1994). This electric field reduced the lifetime of the metastable $2^2S_{1/2}$ state from 0.14 to 1.6×10^{-6} s (through Stark mixing of the $2^2S_{1/2}$ and $2^2P_{1/2}$ states) in accordance with the theoretical predictions of Bethe and Salpeter (1957) which have been experimentally verified by Sellin (1964). A quench field of 15 V cm^{-1} was found to be high enough to quench most of the H(2s) atoms formed in (1), the process of interest, while resulting in only minimal quenching of the faster H(2s) atoms arising from dissociative excitation of the approximately 25% hydrogen molecules present in the target beam.

The 121.6 nm Lyman alpha radiation emitted from the quench field region was viewed at 90° to the quench field direction within a well defined narrow angular range by means of an 18 stage Thorn-EMI 642/2 multiplier fitted with a LiF window. As in our previous work, the transmittance of this window together with the photoelectric emission characteristics of the first dynode provided a wide band filter with an admittance extending from about 104 to 140 nm.

A modulation technique was employed to distinguish the Lyman alpha signals (by virtue of their specific frequency and phase) arising from the process of interest from unwanted signals arising from the interaction of the ion beam with the background gas. In these measurements, the electric field applied to the quenching grids was modulated by a square wave voltage at a frequency of 60 Hz. It is important to note that, since the electric field quenching was carried out beyond the crossed-beam region, fast excited atoms formed by electron capture could not decay within the field of view of our detector. In addition, the slow H(2p) atoms and other short-lived species formed by direct excitation in the crossed-beam region, decayed before entering the field of view of the detector.

2.2. Signal analysis and measurement procedure

As shown in our previous paper (Hughes *et al* 1994), the observed Lyman alpha signal may be expressed in terms of contributions from (1) and from undissociated H_2 molecules present in the target beam. With the discharge turned off when the beam is entirely molecular, the Lyman alpha signal per unit ion beam current is given by

$$S_f = K\sigma_m(2M)^{1/2} \tag{2}$$

where σ_m is the cross section for electric-field-induced Lyman alpha emission following H(2s) formation through dissociative excitation; M is the mass of the hydrogen atom and K is a constant of proportionality. With the discharge on, when the hydrogen beam contains a fraction D of atoms, the Lyman alpha signal per unit ion beam current observed with *the same total mass flow* is given by

$$S_0 = K\sigma_a M^{1/2} 2D + K\sigma_m(2M)^{1/2}(1 - D) \tag{3}$$

where σ_a is the cross section for electric-field-induced Lyman alpha emission from H atoms. Our measurements over the range 10–100 keV showed that the contribution from dissociative excitation was less than 2% of the signal so that we can write

$$\sigma_a \approx S_0/(2KDM^{1/2}) = kS_0 \tag{4}$$

where k is a constant for a particular hydrogen beam with a certain value of D.

Relative cross sections σ_a over the range 10–100 keV were obtained from (4). The constant k was then effectively determined by normalizing these relative cross sections to the previous low-energy value of σ_a measured by Morgan *et al* (1973) at the single energy of 20 keV in the range of overlap between the two sets of data. It is important to note that, the fact that the quench radiation was viewed at 90° in this experiment and in the experiment of Morgan *et al* (1973) obviated the need to correct the present measurements for the anisotropy of the radiation arising from polarization.

As in our recent studies of 2s excitation in He^{2+}–H collisions (Hughes *et al* 1994), we have considered the extent to which cascade contributions from higher states are likely to affect our measured cross sections. The main contribution is from the 3p state and the branching ratio of the 3p to 2s transition is 11.8%. Cross sections for 3p excitation in H^+–H collisions recently measured by Detleffsen *et al* (1994) above 40 keV indicate

Table 1. Cross sections for 2s excitation of H atoms in H^+–H collisions. The reading uncertainties associated with individual values are shown as a standard deviation. All cross sections, which are uncorrected for cascading, are subject to an estimated additional uncertainty of ±30% in absolute magnitude.

Energy (keV)	Cross section (10^{-17} cm^2)
10	0.61 ± 0.10
20	1.05 ± 0.09
30	1.25 ± 0.11
40	1.39 ± 0.08
50	1.32 ± 0.07
60	1.22 ± 0.08
70	1.12 ± 0.07
80	1.04 ± 0.07
90	0.94 ± 0.05
100	0.87 ± 0.07

a cascade contribution increasing from about 8% at 40 keV to 19% at 100 keV. While cascading is clearly significant at the higher energies, as in our previous work (including the measurements of Morgan *et al* (1973)), we have not included any estimated cascade corrections in our measured cross sections shown in table 1. Here random errors are shown as a standard deviation. All the cross sections are subject to an additional estimated uncertainty of ±30% as a consequence of our normalization procedure.

3. Results and discussion

The present cross sections for (1) in the energy range 10–100 keV are shown in figure 1 together with the previous data of Morgan *et al* (1973) and of Chong and Fite (1977) in the respective energy ranges 5–26 and 6–25 keV. It is worth noting that, although the present cross sections were normalized to the 20 keV value of Morgan *et al* (1973), there is good accord between the two sets of data in the energy range of overlap. The much smaller cross sections of Chong and Fite (1977) may partly reflect their use of a different normalization procedure.

The present data exhibit a broad peak in the cross section at about 40 keV and, together with the low-energy values of Morgan *et al* (1973), they also confirm an undulatory energy dependence of the type predicted by some theoretical descriptions. Many of the earlier

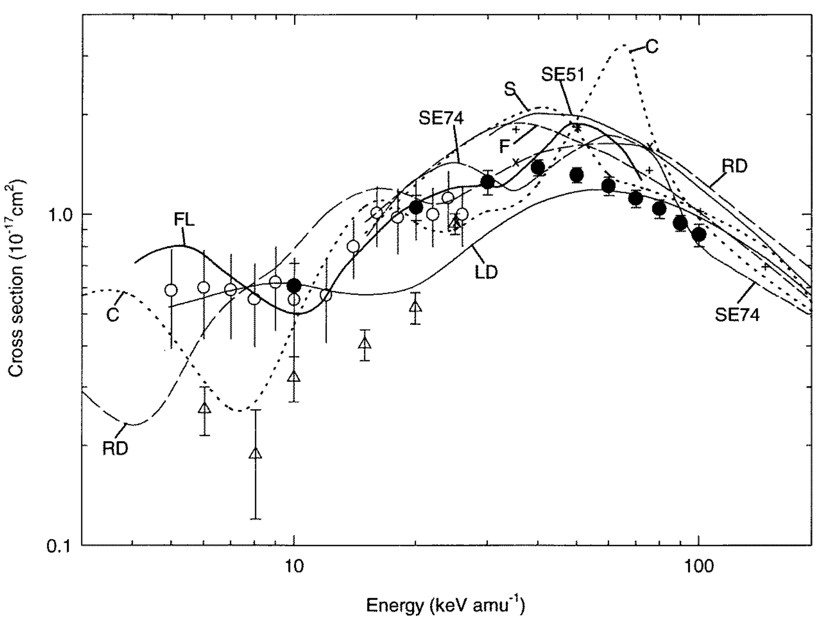

Figure 1. Cross sections for 2s excitation of H atoms in H^+–H collisions. Experiment: (●), present data, (○), Morgan *et al* (1973), (△), Chong and Fite (1977). Theory: Curve FL, Fritsch and Lin (1983), two-centre AO expansion. Curve C, Cheshire *et al* (1970) , seven-state close coupling. Curve RD, Rapp and Dinwiddie (1972), seven-state close coupling. Curve S, Shakeshaft (1978), two-centre hydrogenic expansion. Curve LD, Lüdde and Dreizler (1989), two-centre optical model. Curve F, Ford *et al* (1993), single-centred expansion. Curve SE51, Slim and Ermolaev (1994), two-centre AO 51-state expansion. Curve SE74, Slim and Ermolaev (1994), two-centre AO 74-state expansion. (+), Slim and Ermolaev (1994), two-centre AO 57-state expansion. (×) Slim and Ermolaev (1994), two-centre AO 93-state expansion.

calculations have been discussed by Morgan *et al* (1973) and, more recently, Fritsch and Lin (1991) have reviewed calculations based on the semiclassical close-coupling approach. In figure 1 we include results of a few selected calculations based on models appropriate to the energy range considered.

None of the calculations shown in figure 1 provide a good fit to the present experimental data especially in the region of the cross section maximum where most of the theoretical values are too large. The coupled-state calculations of Fritsch and Lin (1983), which employ a two-centre atomic orbital basis of 40 states and pseudostates with $l \leqslant 1$, while overestimating values in the region of the cross section maximum, probably provide the best overall agreement with the available experimental data down to 5 keV. The earlier seven-state calculations of Cheshire *et al* (1970), which provide a reasonably good general description of both 2s and 2p formation through both excitation and charge transfer below 26 keV (see Morgan *et al* 1973), can be seen to provide a large overestimate of process (1) though a pronounced peak in the region of 65 keV. The Sturmian basis of 70 states with $l \leqslant 2$ employed by Shakeshaft (1978), while in reasonable accord with experiment above about 60 keV, leads to a peak cross section which is too large but at about the correct energy. The seven-state calculations of Rapp and Dinwiddie (1972) can be seen to predict a less pronounced cross section peak but at too high an energy.

In the recent calculations of Slim and Ermolaev (1994) the semiclassical impact-parameter coupled-channel method was used with different choices of basis sets. The first of these (B51) used an asymmetrical basis including only the 1s state of the projectile together with a large set of 50 bound states and pseudostates with $l \leqslant 3$ centred on the target. The second (B74) used a sequence of 37 basis sets centred on both the projectile and target. In further calculations at a few representative energies, the number of states on the target was increased to 56, first in B57 with only the 1s state on the projectile and then in B93 with the 37 projectile states included. In figure 1, the results of the B51 calculations can be seen to be in accord with the present data to the extent that a single broad maximum is predicted at about the right energy although the magnitude of the cross sections is too large. The few points calculated using the B57 basis predict lower values which are in better accord with experiment. In contrast, the B74 basis and the few points calculated with the B93 basis predict structure which is not in accord with experiment. Calculations by Lüdde and Dreizler (1989) using the optical potential model with the instantaneous approximation, can be seen to exhibit an undulatory structure similar to the available experimental data but the theoretical values at intermediate energies are far too small. However, the more recent calculations of Henne *et al* (1993) using the doorway approximation (not shown) are in good agreement with the B51 calculations of Slim and Ermolaev (1994). The calculations of Ford *et al* (1993), which address the problem of convergence of the cross section for a target centred basis, can be seen to predict cross sections close to the B57 values obtained by Slim and Ermolaev (1994).

4. Conclusions

Cross sections for 2s excitation of H atoms in H^+–H collisions have been measured for impact energies in the range 10–100 keV. The results extend previous measurements carried out in this laboratory (Morgan *et al* 1973) in the range 5–26 keV and allow a much needed check of theoretical predictions at intermediate energies where the values obtained are known to be strongly dependent on the choice of basis set. Our measured cross sections, which have been normalized to the measurements of Morgan *et al* (1973) at 20 keV are shown to be consistent with these data in the energy range of overlap. Values, which are roughly a factor of five smaller than the corresponding 2p excitation cross sections, are shown to attain a broad maximum centred on an energy of about 40 keV. A comparison of the available experimental data with a number of representative theoretical predictions shows that, while some of these predict a similar undulatory energy dependence, good quantitative agreement with experiment is limited. However, the present results do confirm the view of Slim and Ermolaev (1994) that, in close-coupling calculations, a large basis set which includes a large number of projectile states at the expense of target states, is less satisfactory at intermediate energies than a basis set which includes only the dominant 1s capture channel.

Acknowledgments

This work forms part of a programme supported by a Rolling Research Grant from the Engineering and Physical Sciences Research Council. One of us (DPH) is indebted to the Department of Education, Northern Ireland for the award of a Research Studentship.

References

Bethe H A and Salpeter E E 1957 *Quantum Mechanics of One and Two Electron Atoms* (New York: Academic)
Cheshire I M, Gallaher D F and Taylor A J 1970 *J. Phys. B: At. Mol. Phys* **3** 813
Chong Y P and Fite W L 1977 *Phys. Rev.* A **16** 933
Detleffsen D, Anton M, Werner A and Schartner K-H 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 4195
Ford A L, Reading J F and Hall K A 1993 *J. Phys. B: At. Mol. Opt. Phys.* **26** 4537
Fritsch W and Lin C D 1991 *Phys. Rep.* **202** 1
——1983 *Phys. Rev.* A **27** 3361
Henne A, Lüdde H J, Toepfer A, Gluth T and Dreizler R M 1993 *J. Phys. B: At. Mol. Opt. Phys.* **26** 3815
Hughes M P, Geddes J and Gilbody H B 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 1143
Lüdde H J and Dreizler R M 1989 *J. Phys. B: At. Mol. Opt. Phys.* **22** 3243
McCullough R W, Geddes J, Donnelly A, Liehr M, Hughes M P and Gilbody H B 1993 *Meas. Sci. Technol.* **4** 79
Morgan T J, Geddes J and Gilbody H B 1973 *J. Phys. B: At. Mol. Phys.* **6** 2118
Rapp D and Dinwiddie D 1972 *J. Chem. Phys.* **57** 4919
Reinhold C O, Olson R E and Fritsch W 1990 *Phys. Rev.* A **41** 4837
Sellin I A 1964 *Phys. Rev.* A **136** 1245
Shakeshaft R 1978 *Phys. Rev.* A **18** 1930
Slim H A and Ermolaev A M 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** L203