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Excitation of the $n=2$ states of H in He^{2+}–H collisions

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Abstract. A crossed beam method has been used to study the excitation of the $n=2$ states of H in He^{2+}–H collisions by observing both spontaneous and electric field induced Lyman alpha radiation. Cross sections for H(2p) and H(2s) formation have been determined in the respective energy ranges 13.3–66.6 keV amu^{-1} and 33.3–66.6 keV amu^{-1} and shown to be in reasonable general accord with the predictions of recent close-coupling calculations.

1. Introduction

A detailed understanding of the process of excitation of H atoms in collisions with He^{2+} ions is directly relevant to schemes for diagnostics of high temperature plasmas and to the modelling of neutral hydrogen beam penetration in large tokamak devices (cf Janev 1989). Reliable experimental data can also provide an important check on the range of validity of theoretical descriptions which are difficult and complex. Close coupling calculations by Fritsch *et al* (1991) for direct $H(n=2)$ and $H(n=3)$ excitation in the range 1–300 keV amu^{-1} predict interesting structures which indicate the complexity of the excitation mechanism arising from competition between the processes of direct excitation and excitation as a result of electron capture.

In previous work in this laboratory (Donnelly *et al* 1991) we used a modulated crossed beam method to study Balmer alpha emission resulting from $H(n=3)$ excitation in He^{2+}–H collisions. In the present work we have used a similar approach to study Lyman alpha emission from spontaneous decay of H(2p) atoms and electric field induced decay of H(2s) atoms formed in He^{2+}–H collisions. Cross sections have been determined for the direct excitation processes

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

and

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

in the energy ranges 13.3–66.6 keV amu^{-1} and 33.3–66.6 keV amu^{-1} respectively.

The only previous experimental studies by Hoekstra and Beijers (cited by Fritsch *et al* 1991) have been confined to (1) in the limited energy range 4–13.3 keV amu^{-1}.

2. Experimental approach

2.1. General description

A momentum analysed beam of $^3He^{2+}$ ions of the required energy after modulation at 60 Hz by electrostatic deflection and collimation, was arranged to intersect at 90° a

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thermal energy beam of highly dissociated hydrogen. The primary ion beam flux was measured directly by means of a screened Faraday cup located beyond the beam intersection region which was maintained at a pressure of at least 3×10^{-6} Torr.

The hydrogen beam was derived from a 2.45 GHz microwave discharge source recently developed in this laboratory (McCullough *et al* 1993). Although capable of providing high intensity beams with dissociation fractions D greater than 0.9, it was necessary to insert a light baffle made from teflon in the exit canal of the atom source to minimise the amount of Lyman alpha radiation from the source reaching the detector. The presence of the light baffle reduced the dissociation fraction D to about 0.75. The dissociation fraction of the hydrogen beam was determined by means of a quadrupole mass spectrometer. A small aperture in line with the beam allowed a small sample to pass into a differentially pumped region where it was modulated by a 180 Hz mechanical chopper prior to analysis by the quadrupole mass spectrometer. The H_2^+ mass spectrometer signal arising from the undissociated component of the beam was recorded with the discharge source on and off to provide signals $S_o(H_2^+)$ and $S_f(H_2^+)$ respectively. The dissociation fraction could then be obtained from the expression.

$$D = 1 - S_o(H_2^+)/S_f(H_2^+). \tag{3}$$

For a constant hydrogen mass flow rate, a change in the temperature of the beam due to the discharge could result in a change in the number density of the beam. However when a He/H_2 mixture was fed into the microwave discharge, the He^+ signal recorded by the mass spectrometer was found to be unchanged when the discharge was switched on. It was concluded that the hydrogen beam temperature was not appreciably changed by the discharge.

The crossed beam intersection region was viewed by an appropriately positioned detector to record 121.6 nm Lyman alpha radiation emitted within a well defined solid angle. The two different arrangements used for studies of (1) and (2) for H(2p) and H(2s) formation are illustrated schematically in figures 1 and 2.

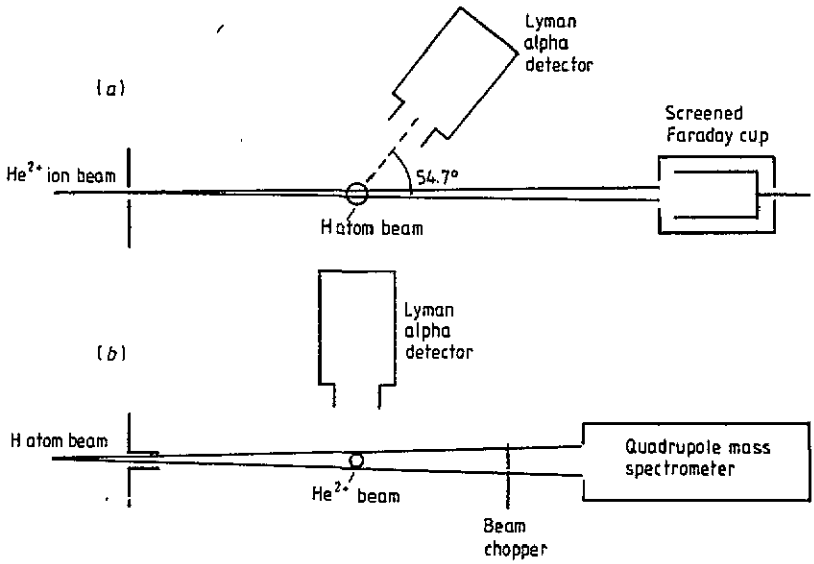

Figure 1. Simplified schematic diagram of experimental arrangement for studies of H(2p) formation in He^{2+}–H collisions viewed (*a*) along the H atom beam axis and (*b*) along the He^{2+} beam axis.

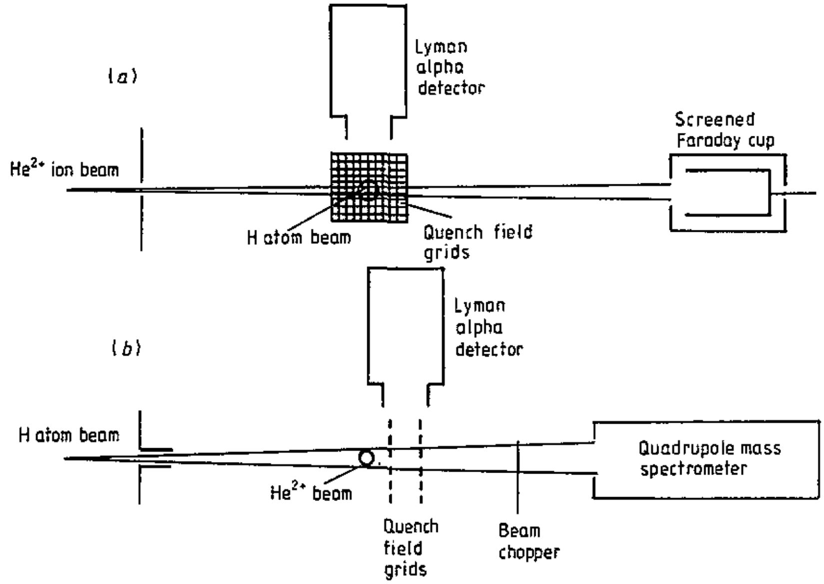

Figure 2. Simplified schematic diagram of experimental arrangement for studies of H(2s) formation in He^{2+}–H collisions viewed (*a*) along the H atom beam axis and (*b*) along the He^{2+} beam axis.

The Lyman alpha detector used in these measurements consisted of an 18 stage EMI 9642 electron multiplier. This was fitted with a LiF window. Earthed grids were placed in contact with each face of the LiF window to limit the build-up of stray surface charge and to restrict field penetration from the multiplier. The transmittance of the LiF window together with the photoelectric emission characteristics of the first dynode provided a wide band pass filter with an admittance extending from about 104 nm to 140 nm. This included the 121.6 nm Lyman alpha emission but excluded all other atomic hydrogen transitions. However, emissions from H_2 bands and from excited products of electron capture had to be considered as described in section 2.2. The angular acceptance of the Lyman alpha detector was limited to the crossed beam intersection region by means of a cylindrical shield.

2.2. *Studies of H(2p) formation*

The H(2p) atoms formed by excitation of the thermal energy H atoms in (1) decay spontaneously with a lifetime of 1.59×10^{-9} s and emit Lyman alpha radiation within the field of view of the detector. In these measurements (figure 1) the Lyman alpha detector was set at 54.7° with respect to the He^{2+} beam in order to obviate the need for a polarization correction (Smit 1935).

The recorded Lyman alpha signal included a component arising from the interaction of the ion beam with the background gas in the interaction region. In order to determine the magnitude of this background correction, the hydrogen beam was turned off and a separate gas feed was used to raise the pressure in the crossed beam chamber to the same value recorded when the hydrogen beam was present.

It was necessary to take account of the contribution to the Lyman alpha signal from undissociated H_2 molecules present in the target beam. With the discharge off, the Lyman alpha signal per unit ion beam current arising from the undissociated hydrogen target beam is given by (Morgan *et al* 1973) the expression

$$S_f = K\sigma_m(\mathrm{Ly})(2M)^{1/2} \tag{4}$$

where $\sigma_m(\mathrm{Ly})$ is the cross section for spontaneous Lyman alpha emission by dissociative excitation of H_2, M is the mass of the hydrogen atom and K is a constant of proportionality. With the discharge on the Lyman alpha signal per unit ion beam current from the target hydrogen beam (with the same total mass flow) is given by

$$S_o = K\sigma(\mathrm{Ly})M^{1/2}2D + K\sigma_m(\mathrm{Ly})(2M)^{1/2}(1-D) \tag{5}$$

where $\sigma(\mathrm{Ly})$ is the cross section for spontaneous Lyman alpha emission from H atoms. From equations (4) and (5) we then obtain

$$\frac{\sigma(\mathrm{Ly})}{\sigma_m(\mathrm{Ly})} = \frac{1}{D\sqrt{2}}\left(\frac{S_o}{S_f} - (1-D)\right). \tag{6}$$

Measurements at each particular energy carried out within the range 13.3–66.7 keV amu^{-1} were based on the average of fifteen sets of readings. Relative values of $\sigma(\mathrm{Ly})/\sigma_m(\mathrm{Ly})$ determined from equation (6) were then normalized by reference to total cross sections for Lyman alpha emission from both direct excitation and charge transfer in H^+–H collisions at 26 keV measured previously in this laboratory (Morgan *et al* 1973). The normalization was carried out by substituting a proton beam of energy 26 keV for the He^{2+} beam under the same experimental conditions and thereby obtaining the relative atomic to molecular emission cross section for H^+ impact. In the normalization, the Lyman alpha emission cross section $\sigma_m^p(\mathrm{Ly})$ for H^+–H_2 collisions measured by Morgan *et al* (1973) could not be used directly since their Lyman alpha detector (unlike the present detector) was able to reject possible emissions from H_2 bands of wavelength near 121.6 nm. The value of $\sigma_m(\mathrm{Ly})$ for He^{2+} impact in equation (6) was obtained from $\sigma_m(\mathrm{Ly}) = (S_f/S_f^p)\sigma_m^p(\mathrm{Ly})$ where S_f and S_f^p were the Lyman alpha signals recorded per unit beam intensity for He^{2+} and H^+ impact respectively under the same experimental conditions. Values of $\sigma(\mathrm{Ly})$ could then be obtained over the full energy range.

It was necessary to estimate to what extent one-electron capture into the $n=4$ sublevels of He^+ followed by the emission of 121.51 nm radiation through 4s–2p, 4p–2s and 4d–2p decay could contribute to the signals recorded by our broad band Lyman alpha detector. Cross sections for capture into the $He^+(n=4)$ states measured by Hoekstra *et al* (1991) and Frieling *et al* (1992) over the present energy range were used in conjunction with the known lifetimes of the $n=4$ sublevels to apply the correction at each energy. The maximum contribution at our lowest energy of 13 keV amu^{-1}, was estimated to be no more than 17% and no greater than 2% at 66.6 keV amu^{-1}.

We also considered the need to correct our measured 2p excitation cross sections to allow for cascade contributions from the $H(n=3)$ sublevels. Estimates were obtained from cross sections for 3s, 3p and 3d excitation calculated by Fritsch *et al* (1991) which are in good accord with our previously measured (Donnelly *et al* 1991) cross sections for Balmer alpha emission. Estimated cascade corrections ranged from about 4% at 13.3 keV amu^{-1} to 7% at 66.7 keV amu^{-1}. However, since these estimated corrections are small and based upon theoretical predictions, they have not been applied to our measurements.

Table 1. Cross sections for H(2p) formation in He^{2+}–H collisions.

Energy (keV amu^{-1})	Cross section (10^{-16} cm^2)
66.7	1.34 ± 0.20
60.0	1.19 ± 0.01
53.3	1.26 ± 0.07
46.7	1.20 ± 0.08
40.0	1.19 ± 0.19
33.3	1.32 ± 0.13
26.7	1.28 ± 0.11
20.0	0.88 ± 0.13
13.3	0.30 ± 0.15

Cross sections for 2p excitation are given in table 1 where random errors are shown as a standard deviation. Our normalization procedure gives rise to an additional estimated uncertainy of ±30%.

2.3. Studies of H(2s) formation

In the studies of H(2s) formation an electric field of up to about 100 V cm^{-1} was applied to the H atom beam at a point 1 cm beyond the crossed beam region (figure 2) by means of a pair of grids. In this way, the lifetime of the metastable $2^2S_{1/2}$ state was reduced by electric field quenching (through Stark effect mixing of the $2^2S_{1/2}$ and $2^2P_{1/2}$ states) from 0.14 s to about 3×10^{-8} s in accord with the theoretical values of Bethe and Salpeter (1957) which have been experimentally verified by Sellin (1964). It was found advantageous to use a quench field of about 15 V cm^{-1} which was high enough to quench most of the H(2s) atoms formed by direct excitation of the thermal energy H atoms but provided only minimal quenching of the faster H(2s) atoms arising from dissociative excitation of the H_2 molecules in the target beam.

The signal analysis followed a procedure similar to that described in section 2.2. Measured relative cross sections in the range 33.3–66.7 keV amu^{-1} were again determined by normalization to the total cross sections for H(2s) formation in H$^+$–H collisions measured previously by Morgan *et al* (1973) by substitution of a proton beam at 26 keV amu^{-1}. It should be noted that since the Lyman alpha quench radiation was viewed at 90° with respect to the primary ion beam both in the present experiment and in the measurements of Morgan *et al* (1973) a correction to take account of the polarization of the quench radiation was unnecessary.

A correction of not more than 7% was estimated but not applied to the measured cross sections to allow for cascading from the 3p state believed to be the main source of cascading. This was estimated using the populations of the $n = 3$ sublevels calculated by Fritsch *et al* (1991).

Cross sections for 2s excitation are given in table 2 where the random errors are shown as a standard deviation. As in the case of our 2p cross sections, an additional estimated uncertainty of ±30% arises from our normalization procedure.

3. Results and discussion

Our measured cross sections for the 2p excitation process (1) in the range 13.3–66.7 keV amu^{-1} are shown in figure 3. The low energy data due to Hoekstra and

Table 2. Cross sections for H(2s) formation in He^{2+}–H collisions.

Energy ($keV\,amu^{-1}$)	Cross section ($10^{-16}\,cm^2$)
66.7	5.16 ± 1.20
60.0	5.59 ± 1.09
53.3	6.25 ± 0.98
46.7	5.03 ± 0.92
40.0	3.51 ± 0.58
33.3	3.28 ± 1.45

Beijers (cited by Fritsch *et al* 1991) in the range 5.3–13.3 $keV\,amu^{-1}$ are also shown for comparison together with theoretical predictions by Fritsch *et al* (1991), by Bransden and Noble (1981) by Krstic and Janev (1993) and by Lundsgaard and Nielsen (1993) within the range 3–100 $keV\,amu^{-1}$. The two sets of experimental data can be seen to be in satisfactory general accord to the extent that, at the single common energy of 13.3 $keV\,amu^{-1}$, agreement is within the combined limits of uncertainty.

The semiclassical close coupling calculations of Fritsch *et al* (1991) below 10 $keV\,amu^{-1}$ employed a set of two-centre atomic basis states that included united atom orbitals as pseudostates at the two centres (AO + basis). The basis set of hydrogenic states included 31 centred on the H atom and 23 centred on the He nucleus. The calculations above 10 $keV\,amu^{-1}$ employed a basis set which included the dominant atomic bound states of the colliding systems augmented with some bound and positive-energy pseudostates on the atomic centres. The two sets of measured 2p excitation cross sections, while not in close numerical agreement with the values predicted by Fritsch *et al* (1991), do exhibit similar general features. However, below 13.3 keV where there is evidence of structure, a detailed comparison is precluded by the large experimental uncertainties.

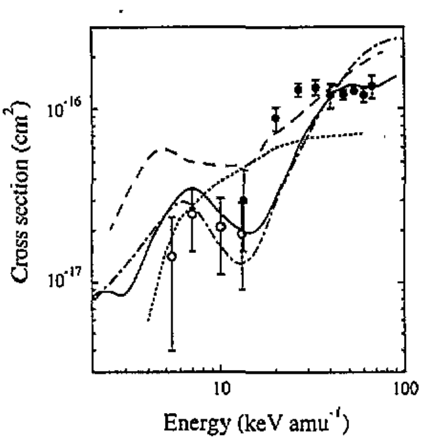

Figure 3. Cross sections for H(2p) formation in He^{2+}–H collisions. ●, Present data; ○, Hoekstra and Beijers (cited by Fritsch *et al* 1991); ——, theory, Fritsch *et al* (1991); – – –, theory, Bransden and Noble (1981); — — —, theory, Lundsgaard and Nielsen (1993); - - - -, theory, Krstic and Janev (1993).

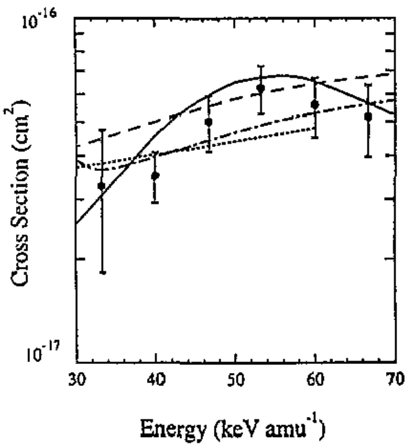

Figure 4. Cross sections for H(2s) formation in He^{2+}–H collisions. ●, present data; ——, theory, Fritsch *et al* (1991); – – –, theory, Bransden and Noble (1981); — — —, theory, Lundsgaard and Nielsen (1993); - - - -, theory, Krstic and Janev (1993).

The recent close-coupling calculations by Lundsgaard and Nielsen (1993) are based on an impact-parameter approach using 14 ETF-modified atomic states. Their values (figure 3), like those of Fritsch *et al* (1991), predict structure which is in reasonable general accord with experiment although at the highest energies considered the agreement is less satisfactory. Bransden and Noble (1981) used a coupled channel calculation which employed the impact parameter formalism with plane wave momentum transfer factors in a two centre atomic expansion with eight terms. For energies up to about 13 keV amu^{-1} the results of these calculations (figure 3), while larger, can also be seen to exhibit some structure. However above 40 keV amu^{-1}, the cross sections calculated by Bransden and Noble (1981) continue to rise while those due to Fritsch *et al* (1991) exhibit structure in better general accord with the present experimental data.

The calculations due to Krstic and Janev (1993) are based on the concept of hidden adiabatic energy crossings and employ about 160 molecular states coupled by 220 radial transitions and rotational transitions. However while their calculated 2p cross sections (figure 3), appear to provide the best fit to the experimental data below about 7 keV amu^{-1}, at higher energies they exhibit a different energy dependence which is devoid of structure and in poor accord with measured values.

Figure 4 shows our measured cross sections for the 2s excitation process (2) in the range 33.3–66.7 keV amu^{-1} compared with the calculated values by Fritsch *et al* (1991), by Bransden and Noble (1981), by Krstic and Janev (1993) and by Lundsgaard and Nielsen (1993). The results of Fritsch *et al* (1991) can be seen to be in good general accord with the present data within the limits of experimental uncertainty. Although the cross sections calculated by Bransden and Noble (1981), by Krstic and Janev (1993) and by Lundsgaard and Nielsen (1993) do overlap the experimental values the predicted energy dependence is less satisfactory over the range shown.

Errea *et al* (1992) have calculated total cross sections for $n=2$ excitation in the velocity range 0.2–2.8 au by the use of the common translation factor method to augment a molecular orbital approach in a semiclassical close coupling calculation. Their cross sections (not shown) are in reasonable accord with the sum of the 2s and 2p for excitation calculated by Fritsch *et al* (1991).

4. Conclusions

The cross sections for 2p and 2s excitation of H in He^{2+}–H collisions measured in this work provide a useful assessment of recent close coupling calculations. In particular, our 2p excitation cross sections together with the lower energy measurements of Hoekstra and Beijers (cited by Fritsch *et al* 1991) exhibit evidence of structure and vary greatly with energy in a manner which is in general accord with the theoretical predictions of Fritsch *et al* (1991). Our measured 2s excitation cross sections also provide support for the close coupling calculations of Fritsch *et al* (1991).

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