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LETTER TO THE EDITOR

New low-energy measurements and calculation of ionization in H^+–H collisions

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Abstract. In response to the strong current theoretical interest in models of low-energy ionization in H^+–H collisions, a crossed-beam coincidence counting technique previously used in this laboratory to obtain cross sections in the range 9–1500 keV amu^{-1} has been successfully adapted to extend the measurements downwards in energy from 9 to 1.25 keV amu^{-1}. In addition, we have extended the triple-centre calculations of McLaughlin B M, Winter T G and McCann J F 1997 *J. Phys. B: At. Mol. Opt. Phys.* **30** 1043, down to 0.5 keV amu^{-1} where agreement with experiment is found to be satisfactory. Our new measurements and calculation are at variance with relative cross sections measured by Pieksma M, Ovchinnikov S Y, van Eck J, Westerveld W B and Niehaus A 1994 *Phys. Rev. Lett.* **73** 46 in the range 1–6 keV amu^{-1}, but are in good accord with their calculations based on the hidden-crossings model including only S and T promotion. However, our measurements and calculation do not support the more recent calculations by Pieksma M, Ovchinnikov S Y and Macek J H 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 1267 in which a contribution from a radial decoupling mechanism is also included in the model.

Accurate cross sections for the simple ionization process

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

over a wide energy range are essential for reliable modelling of plasmas in fusion energy devices and in astrophysical applications. Reliable experimental benchmarks are also valuable for checking the range of validity of the sometimes greatly divergent theoretical predictions based on different models of ionization. The present investigation has been stimulated by a recent experimental study of ionization in H^+–H collisions in the range 1–6 keV amu^{-1} by Pieksma *et al* (1994, 1996) and attempts (Pieksma and Ovchinnikov 1991, Ovchinnikov *et al* 1997, Pieksma *et al* 1994, 1996, 1998) to obtain a satisfactory description of (1) at low keV impact energies in terms of variants of the hidden-crossings model of ionization and including, more recently, a radial decoupling mechanism.

In previous work in this laboratory (Shah and Gilbody 1981, Shah *et al* 1987a) we used a crossed-beam technique incorporating time-of-flight spectroscopy and coincidence counting of the collision products to obtain cross sections for (1) over the overlapping energy ranges 38–1500 and 9–75 keV amu^{-1} with an absolute accuracy estimated to be within ±3.8% and 5%, respectively. These data, together with our corresponding measurements of cross sections for ionization of H(1s) atoms by electron impact (Shah *et al* 1987b), when normalized to theoretical values predicted by the first Born approximation above 1000 keV amu^{-1}, have been shown to be fully consistent with the absolute cross sections for the charge transfer process

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

measured by McClure (1966) at lower energies where cross sections for (2) begin to exceed those for (1).

Our previously measured cross sections for (1) obtained in this way have provided important tests of theory over a wide energy range (cf the review by Gilbody 1994). At the lowest energies considered down to 9 keV amu^{-1}, the calculations of Pieksma and Ovchinnikov (1991) based on the hidden-crossings method were found to provide a good description of our experimental values. These calculations indicate that the saddle-point mechanism contributes significantly to ionization. Both CTMC calculations and triple-centre close-coupling calculations have also indicated the possibility of electrons being balanced by the attractive forces of the two colliding nuclei becoming in effect stranded on top of the internuclear barrier—the saddle point. An electron which stays localized in this region until after the collision will no longer be bound to the nuclei, resulting in ionization. In the recent experimental study of ionization in H^+–H collisions carried out by Pieksma *et al* (1994, 1996) in the range 1–6 keV using a crossed-beam approach, a study of the velocity distributions of the ejected electrons was claimed to provide strong evidence of a saddle-point ionization mechanism above 4 keV and relative cross sections for ionization were determined by integrating the observed electron energy distributions.

In the present work, in order to try to obtain a reliable assessment of theory at low keV energies, we have adapted the crossed-beam coincidence counting technique used in our previous studies (Shah and Gilbody 1981, Shah *et al* 1987a) and managed to extend our cross section measurements from the previous low-energy limit of 9 keV amu^{-1} down to 1.25 keV amu^{-1} by the use of both H^+ and D^+ ion beams. Full details of the basic experimental approach and the signal analysis procedure have been given in our previous papers and only a brief summary of the essential features need be given here.

A well defined momentum-analysed beam of either H^+ and D^+ ions, at an energy adjustable within the range 1.25–9 keV amu^{-1}, was arranged to intersect (at right angles) in a high-vacuum region a thermal energy beam of highly dissociated hydrogen obtained from a tungsten tube furnace source. Slow ions and electrons formed as collision products were extracted by a transverse electric field and counted separately by particle multipliers. Product H^+ ions could be separately distinguished from H_2^+ ions and ions arising from background gas species by their characteristic times of flight to the multiplier. The H^+ ion signal arising from the ionization process (1) could be distinguished from the (up to three orders of magnitude larger) signal arising from the charge transfer process (2) by counting the H^+ ions in coincidence with the electrons arising from the same collision events.

As in our previous work, accurate allowance was made for the contribution to the e^-–H^+ coincidence signal from dissociative ionization of any H_2 molecules present in the target beam and in the background gas. In the present low-energy measurements, particular care had to be taken to ensure that the transverse electric field applied across the beam intersection region was high enough to ensure complete collection of ions and electrons without causing significant deflection of the primary beam and consequent changes to the effective collision volume. As in our previous work (Shah *et al* 1987a) the transverse electric field was applied between two high-transparency grids set into inverted conical electrodes (as in figure 1 of Shah and Gilbody (1982)) so that the extraction field was high only over a limited spatial region where the beams overlap. It was found that, with a primary H^+ beam

Table 1. Cross sections for the ionization of H atoms by H^+ or D^+ ions.

Energy (keV amu^{-1})	Cross sections (cm^2 $\times$ 10^{-18})
1.25	0.39 ± 0.10
1.5	0.55 ± 0.14
2.0	0.75 ± 0.17
2.5	1.38 ± 0.27
3.0	1.68 ± 0.26
4.0	3.2 ± 0.5
5.0	4.7 ± 0.8
6.0	5.8 ± 1.1
7.0	9.6 ± 1.8
8.0	11.3 ± 2.2
9.0	15.5 ± 2.8

of diameter 1 mm intersecting a hydrogen beam of 6 mm diameter, an extraction field of 100 V cm^{-1} produced a negligible change to the effective collision volume. Relative cross sections for (1) obtained from the measured e^-–H^+ coincidence signals for the same target beam conditions at different primary ion energies were then normalized to our previously measured cross sections (Shah *et al* 1987a) at 9 keV.

The results shown in table 1 include individual uncertainties which reflect the degree of reproducibility in terms of experimental parameters and statistical fluctuations assessed at the 67% confidence level. All values are subject to an additional estimated uncertainty of $\pm$8% in absolute value associated with our overall normalization procedure.

The present cross sections in the range 1.25–9 keV amu^{-1} are shown in figure 1 together with some of our previously measured values (Shah *et al* 1987a) down to 9 keV amu^{-1}. We also include for comparison the relative cross sections measured by Pieksma *et al* (1994, 1996) in the range 1–6 keV which, for consistency, have been normalized by extrapolation to our previous measurements (Shah *et al* 1987a). Their cross sections can be seen to decrease much less rapidly with decreasing energy than our values. Comparison can also be made with a number of theoretical predictions shown in figure 1. Calculations by Pieksma *et al* (1994) based on the hidden-crossings model can be seen to provide quite a good overall description of our experimental results. These calculations include contributions from T-type crossings connected with the saddle-point ionization mechanism and S-type crossings associated with the transition from quasi-molecular to united-atom behaviour. In their work, additional low-energy contributions from a radial decoupling mechanism proposed by Ovchinnikov and Macek (1993) involving decoupling of electron and nuclear motion within the united-atom limit were also considered. These contributions were recalculated in a very recent paper (Pieksma *et al* 1998) and the values obtained with the contributions from this radial decoupling mechanism included (figure 1), while in better agreement with the measurements of Pieksma *et al* (1994, 1996), can be seen to decrease much less rapidly than the present experimental values.

At the very low impact energies considered in the present work, values calculated by Bandarage and Parson (1990) based on the classical trajectory Monte Carlo (CTMC) method are, as might be expected, much smaller and diverge increasingly from our measured cross sections with decreasing energy. In the present work we have also extended the recent calculations of McLaughlin *et al* (1997) based on a multi-state close-coupling approach using a triple-centre basis downwards in energy to obtain values in good agreement with the triple-centre calculations of Winter and Lin (1984). Both these

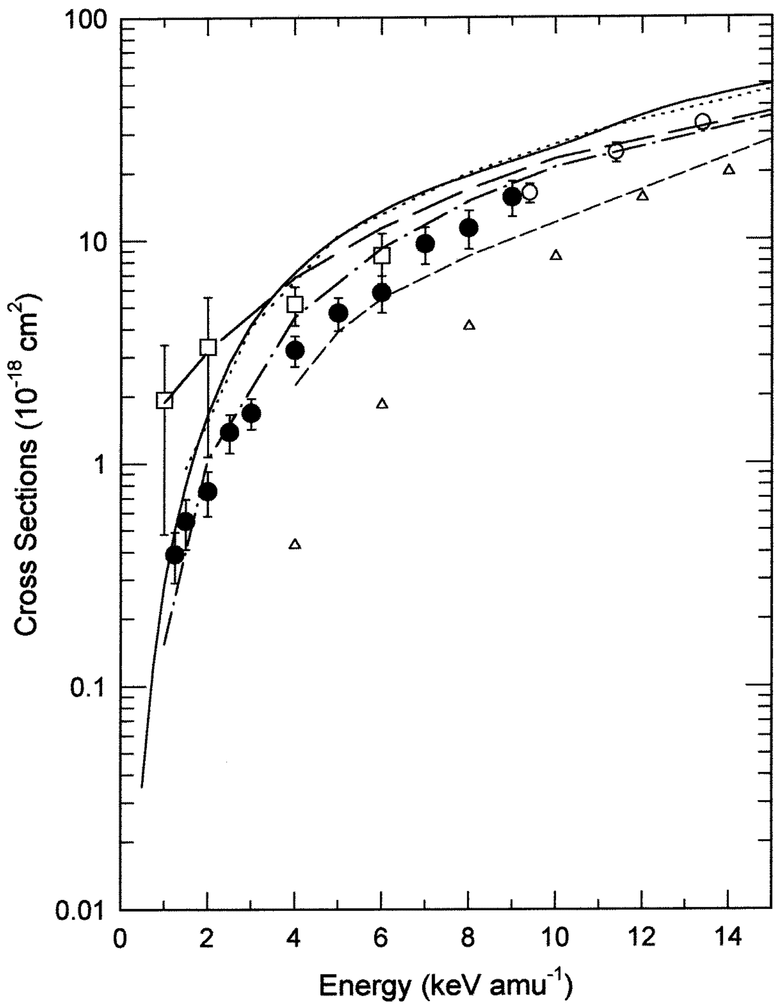

Figure 1. Cross sections for ionization in H^+–H collisions: ●, present cross sections; ○, our previous cross sections (Shah *et al* 1987a); □, relative cross sections measured by Pieksma *et al* (1994, 1996) normalized by extrapolation to values of Shah *et al* (1987a). — · —, hidden-crossing theory including only S and T promotion mechanisms (Pieksma *et al* 1994); — — —, hidden-crossing theory with S and T and radial decoupling mechanisms (Pieksma *et al* 1998); ——, present close-coupling triple-centre calculations; · · · · · ·, close-coupling triple-centre calculations (Winter and Lin 1984); – – –, two-centre close-coupling calculations (Fritsch and Lin 1983). △, CTMC calculations (Bandarage and Parson 1990).

calculations predict values larger than our experimental cross sections, but the discrepancy is smallest at the lowest energies considered. Cross sections calculated by Fritsch and Lin (1983) using a two-centre close-coupling method which extend down to 4 keV amu^{-1}, while smaller, also exhibit closer agreement with experiment at the lowest energies considered.

The velocity scaling of the measured ionization cross sections at low energies is also of interest since this provides an indication of the main mechanism of ionization. Pieksma *et al* (1998) have suggested that an adiabatic description, which predicts cross sections near

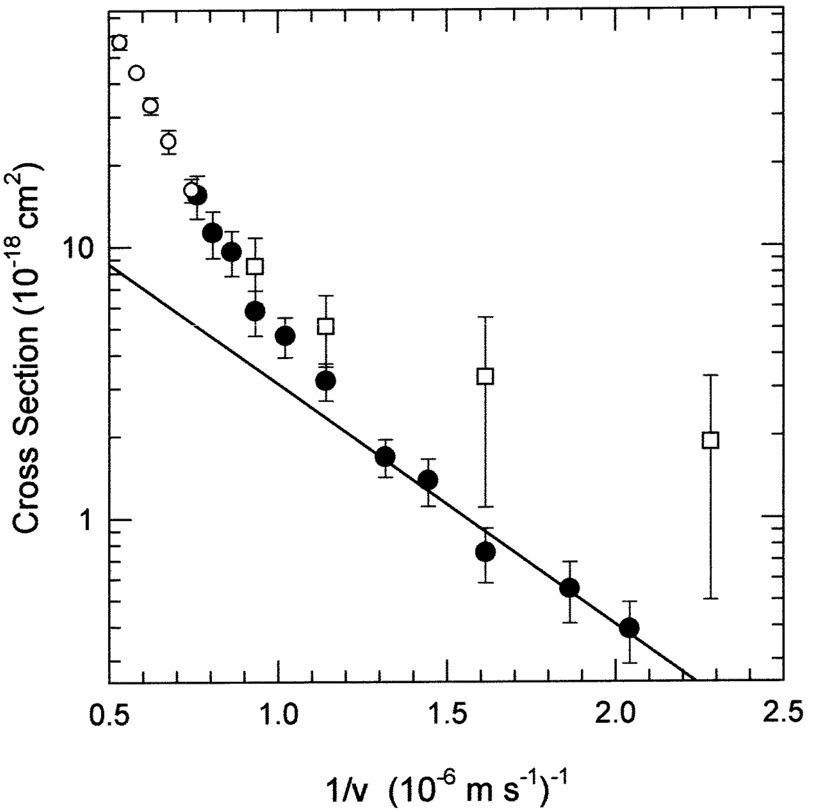

Figure 2. Experimental cross sections for ionization in H^+–H collisions fitted to the relation $\sigma = A\exp(-C/v)$ at low energies (see text). ●, present cross sections; □, relative cross sections measured by Pieksma *et al* (1994, 1996) normalized by extrapolation to values of Shah *et al* (1987a).

threshold scaling with velocity according to a relation of the type

$$\sigma \propto \exp(-2\Delta/v) \tag{3}$$

(where Δ is a constant sometimes called the Massey parameter), is appropriate for the S and T promotion mechanisms in the hidden-crossings model. However, a v^2 dependence might be expected if only the radial decoupling mechanism is significant at sufficiently low velocities. In figure 2 the relation $\sigma = A\exp(-C/v)$ where $A = 23 \times 10^{-18}$ cm^2 $\pm$ 35% and $C = 2.0 \times 10^6$ m s^{-1} $\pm$10% can be seen to provide a good fit to our cross sections below 3 keV amu^{-1} (where the S mechanism is dominant) in accord with the simple adiabatic description. Our results also indicate that $\Delta = 0.46$ au, a value which compares favourably with $\Delta = 0.53$ au predicted by the calculations of Pieksma and Ovchinnikov (1991) for the S mechanism of ionization.

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