One-electron capture by slow Al^{2+} ions in atomic and molecular hydrogen

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Abstract. Cross sections for one-electron capture by Al^{2+} ions in both atomic and molecular hydrogen have been measured and calculated at energies within the range 10–2000 eV amu⁻¹. Measurements are based on the use of a tungsten tube furnace to provide a target of highly dissociated hydrogen. Translational energy spectroscopy used with a specially developed recoil source of metallic ions is used to show that ground-state Al^+ ion production is predominant. The calculations, which are based on a model potential description of the orbitals, predict cross sections which are somewhat larger than our measured values.

I. Introduction

In this paper, we report on a series of experimental and theoretical investigations of charge transfer in Al^{2+}–H and Al^{2+}–H_2 collisions for ion energies in the range 10–2000 eV amu⁻¹.

The Al^{2+}–H system, in particular, is of special interest for a number of reasons. Firstly, since electron capture is expected to take place primarily into the $Al^+(3s^2)^1S$ ground state, the inverse reaction (charge transfer ionisation of Al^+ by H^+) can affect the ionisation equilibrium in laboratory and astrophysical plasmas under certain conditions. This phenomenon is fairly common for metallic ions. For example, Baliunas and Butler (1980) showed how charge transfer ionisation affected the abundance of Si ions in some astrophysical plasmas at temperatures of 2×10^4–10^5 K. Secondly, it may be remarked that whereas the curve crossing responsible for charge transfer in Al^{2+}–H collisions, which is located at around 6.3 a_0, tends to be of adiabatic character, the crossing for Al^{2+}–H_2 collisions around 8 a_0 tends to be diabatic at least in the energy range investigated experimentally here. This fortunate coincidence, together with unfavourable spin weighting factors leading to anomalously small cross sections, make for a critical test of the theoretical model.

The present work is complementary to that of Phaneuf *et al* (1985). However, their experimental measurements for Al^{2+}–H were limited to an upper limit estimate at the single-ion impact at an energy of 23.9 eV amu⁻¹ where the cross section is very small. Their theoretical model for Al^{2+}–H is more detailed (and in principle more accurate) than ours, since they have used a configuration interaction calculation involving all

possible configurations arising from the 3s, 3p and 3d orbitals of Al and the 1s, 2s and 2p orbitals of H. In our work, we shall limit ourselves to a model potential description of the orbitals as in Gargaud *et al* (1989). This method has worked well for more highly charged systems when capture occurs via excited states. It is therefore of some interest to assess the accuracy of the method for the more difficult case of doubly charged ions. In addition, the model potential method can easily be applied to the Al^{2+}-H_2 system, for which the absolute measurements should be the most reliable.

2. Experimental approach

Measurements of the total one-electron capture cross section for Al^{2+} in H and H_2 were carried out using an experimental approach used previously in Belfast (cf McCullough *et al* 1979). In a second experiment, we have used the technique of translational energy spectroscopy (cf McCullough *et al* 1987) to identify the final excited states of the Al^+ product ions. Both experiments made use of a tungsten tube furnace to provide targets of highly dissociated hydrogen.

2.1. Total cross section measurements

The apparatus and measurement procedure have been described in detail previously (Nutt *et al* 1978a, b).

Al^{2+} ions derived from a universal ion source (Hill and Nelson 1965) were accelerated to 8 keV, momentum analysed by a 15° magnetic sector field and subsequently decelerated or accelerated to the required impact energy in the range 5–30 keV. Beam intensities in the range 10^{-10}–10^{-11} A were obtained in the target chamber after electrostatic focusing and collimation.

The Al^{2+} beam was directed into a differentially pumped chamber via a 1 mm entrance aperture where it passed along the axis of a 7.5 cm long 5 mm diameter tungsten tube furnace before leaving via a 6 mm exit aperture. The furnace was symmetrically positioned in the chamber at a distance of 4 mm from the entrance and exit apertures. The beam emerging from the target region was passed into a detection chamber where the Al^+ product ions and the Al^{2+} primary ions where separated by electrostatic deflection and simultaneously recorded as currents to Faraday cups.

The furnace operating procedure was the same as that used previously (Nutt *et al* 1978a, b). The ratio S_c of the Al^+ product ion current to the Al^{2+} primary beam current was determined with the furnace at 1400 K when the hydrogen target was completely molecular. The corresponding ratio S_H was obtained in the furnace at 2400 K when only a fraction f of the H_2 molecules were undissociated. The ratio $\sigma_{21}(H)/\sigma_{21}(H_2)$ of the Al^+ formation cross sections in H and H_2 is given by

$$\frac{\sigma_{21}(H)}{\sigma_{21}(H_2)} = \left(\frac{S_H}{S_c} - f\right)\frac{N_m}{N_a} \tag{1}$$

where N_m is the number density of H_2 at 1400 K and N_a is the number density of H at 2400 K.

The fraction f was determined as in our previous work (cf Nutt *et al* 1978a) from measurements of H^- production by two-electron capture by protons in single collisions with the undissociated H_2. The ratio N_m/N_a was obtained as also described in our previous work (Shah and Gilbody 1978) by reference to the ratio $\sigma_{10}(H)/\sigma_{10}(H_2)$ of

the total and electron capture cross sections for protons in H and H_2 measured by McClure (1966) with an accuracy believed to be within 3%. Absolute values of $\sigma_{21}(H_2)$ were obtained by operating the furnace at room temperature and calibrating the H_2 target thickness by reference to the cross section $\sigma_{10}(H_2)$ for 15 keV H^+ in H_2 measured by Stier and Barnett (1956) with an accuracy believed to be within 5%. Equation (1) was then used to obtain $\sigma_{21}(H)$ from the measured ratios $\sigma_{21}(H)/\sigma_{21}(H_2)$. Our normalisation procedure is estimated to provide an overall uncertainty of 11% in the absolute magnitude of $\sigma_{21}(H)$.

2.2. Translational energy spectroscopy

The basic method, developed previously in Belfast for measurements in atomic hydrogen, was used to identify and quantify the excited Al^+ product channels in both Al^{2+}-H and Al^{2+}-H_2 collisions. Studies of the difference ΔT in kinetic energy between the primary Al^{2+} ion and forward scattered Al^+ product ions allow the energy defects $\Delta E \simeq \Delta T$ of the relevant collision channels to be identified. The basic apparatus and measuring procedure differed from that used previously (cf McCullough *et al* 1987) only in the use of a recoil ion source developed specially for metallic species. A 5 kW electron beam evaporator was used to produce aluminium vapour which was then bombarded by a beam of Ar ions from a van de Graaff accelerator operating at 1.5 MV. The resulting slow multiply charged recoil Al ions (characterised by a small energy spread) were then used as primary ions in our translational energy spectroscopy apparatus.

A schematic diagram of the metallic vapour recoil source in shown in figure 1. A sample of pure aluminium was mounted on a tantalum hearth maintained at up to +10 kV and heated by electrons derived from a ring-shaped filament. An electron current of up to 100 mA at 6 keV was typical. The hearth was adjustable in height and was positioned about 35 mm below the axis of the Ar ion beam from the van de Graaff generator. Screening grids were used to prevent penetration of electric fields from the evaporator into the extraction region where transverse fields of less than 1 V cm^{-1} were used to direct the Al recoil ions into the main apparatus. The recoil source was also
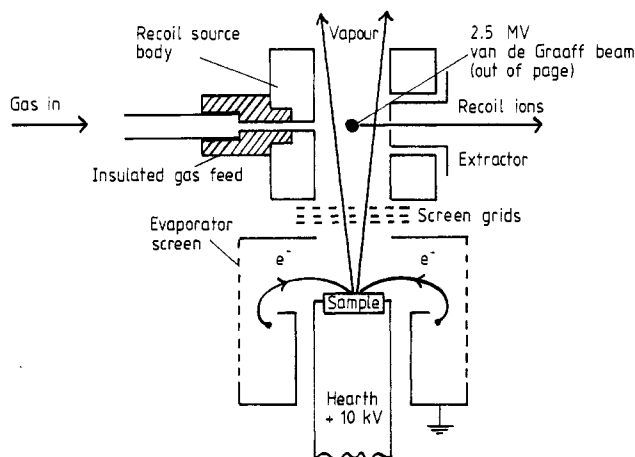

Figure 1. Schematic diagram of metallic vapour recoil ion source used in translational energy spectroscopy measurements.
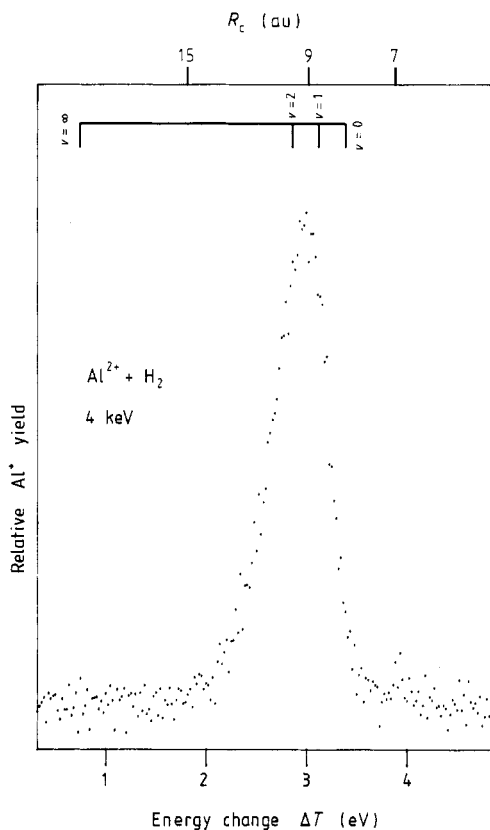
Figure 2. Energy change spectrum for one-electron capture by 4 keV Al^{2+} ions in H$_2$.

provided with a gas feed tube so that when required (e.g. for calibration measurements) a simple gas target could be substituted for the metallic vapour target.

A typical energy-change spectrum obtained from the translation energy spectrometer is shown in figure 2. These measurements for 4 keV Al^{2+} in H$_2$ will be discussed in § 4.

3. Theoretical methods

The theoretical calculations are based on the methods first tested by Benmeuraiem *et al* (1987) for Ar^{6+}–He and later developed by Gargaud *et al* (1989) and Gargaud and McCarroll (1989) to study O^{3+}–H and Ne^{3+}–H. A suitably optimised model potential is used to generate the molecular orbitals which are involved in the charge transfer process. The method of Grice and Herschbach (1974) is then used to construct the diabatic wavefunctions and the corresponding coupling matrix elements required for the collision dynamics. In this way, the effects of electron correlation are correctly taken into account in the description of the asymptotic channel states. On the other hand, the effects of correlation on the dynamical transfer from one centre to the other are neglected (cf McGuire 1987).

Translation effects are taken into account by the introduction of appropriate reaction coordinates in a quantum mechanical formulation of the collision dynamics (Gargaud *et al* 1987).

It should be noted that spin conservation in the collision leads to important spin-selective effects. In the case of Al^{2+}-H, where the asymptotic entry channel states are correlated to both a $^1\Sigma$ and $^3\Sigma$ molecular state of (AlH)$^{2+}$, only the $^1\Sigma$ state is susceptible to interaction with the exit channels Al$^+$(3s^2)^{1}S or Al$^+$(3s, 3p)^{1}P. The cross sections for capture must therefore be weighted by a factor of $\frac{1}{4}$ to take account of the fact that, on average, only 25% of the entry channel states follow the $^1\Sigma$ molecular state. In the case of Al^{2+}-H$_2$, where once again only the Al$^+$(3s^2)^{1}S state is significantly populated, the spin weighting factor is $\frac{1}{2}$. (In this case, if we recouple the spin of the Al$^+$ electron with one of the H$_2$ electrons, there is a 50% probability of forming a singlet or triplet state.) These unfavourable spin-weighting factors explain why the cross sections are anomalously small.

4. Results and discussion

Our experimental and theoretical values of cross sections for one-electron capture by Al^{2+} in H and H$_2$ are given in table 1.

Table 1. Experimental and theoretical total cross sections σ_{21}(H) and σ_{21}(H$_2$) for one-electron capture by Al^{2+} in H and H$_2$.

Al^{2+} energy (keV)	Experiment		Theory	
	σ_{21}(H) (10^{-16} cm^2)	σ_{21}(H$_2$) (10^{-16} cm^2)	σ_{21}(H) (10^{-16} cm^2)	σ_{21}(H$_2$) (10^{-16} cm^2)
5.0	1.97 ± 0.31	4.89 ± 0.32	2.88	9.82
6.0	2.76 ± 0.16	5.14 ± 0.18	3.30	9.58
7.0	2.78 ± 0.36	4.88 ± 0.41	3.67	9.35
8.0	3.32 ± 0.32	5.11 ± 0.20	3.98	9.18
9.0	3.35 ± 0.40	5.07 ± 0.31	4.24	9.04
10.0	3.51 ± 0.28	5.05 ± 0.54	4.46	8.93
11.0	3.44 ± 0.29	4.79 ± 0.15	4.65	8.86
12.5	3.48 ± 0.42	4.68 ± 0.27	4.87	8.76
14.0	3.96 ± 0.51	5.12 ± 0.28	5.06	8.68
15.0	3.46 ± 0.34	4.98 ± 0.27	5.16	8.63
17.5	3.95 ± 0.47	5.02 ± 0.36	5.34	8.54
20.0	4.03 ± 0.32	5.22 ± 0.27	5.46	8.47
25.0	4.59 ± 0.57	5.50 ± 0.20	5.56	8.38
30.0	4.38 ± 0.60	5.22 ± 0.24	5.53	8.29

In figure 3 the present results for Al^{2+}-H collisions are shown. Our experimental values also include the uncertainties associated with the lack of reproducibility of individual cross sections. We also include the previous theoretical values of Phaneuf *et al* (1985). Their single experimental measurement at 23.9 eV amu^{-1} is not shown since this provides only an upper limit estimate of 10^{-16} cm^2 for the cross section. However, their coupled channel calculations are resonably consistent with the present theoretical estimates over a wide energy range. Unfortunately, it is difficult to ascertain whether the difference between the two sets of results is significant since translation effects, which become increasingly important with collision energy, have not been considered in the same way. Comparison with experiment shows that both sets of calculations provide results which are too large by up to 40%.
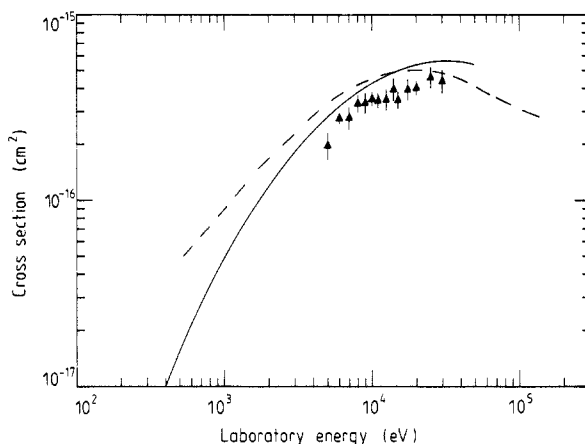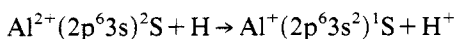
Figure 3. Cross sections for one-electron capture by Al^{2+} in atomic hydrogen. The symbols denote: ▲, present measurements; ——, present calculations; – – –, calculations of Phaneuf *et al* (1985).

Phaneuf *et al* (1985) claim that capture to the $Al^+(3s3p)^1P$ state contributes up to 25% of the total cross section while our theory predicts that this channel is unimportant. Moreover, our translational energy spectrscopy studies provide energy change spectra for Al^{2+}-H and Al^{2+}-H_2 which exhibit only single peaks (an example for Al^{2+}-H_2 is shown in figure 2) consistent with the following predominant channels

$$Al^{2+}(2p^63s)^2S + H \rightarrow Al^+(2p^63s^2)^1S + H^+$$

and

$$Al^{2+}(2p^63s)^2S + H_2 \rightarrow Al^+(2p^63s^2)^1S + H_2^+(^2\Sigma_g)$$

in which ground-state $Al^+(^1S)$ ions are formed. In addition the spectrum for Al^{2+}-H_2 (see figure 2) indicates that H_2^+ is formed primarily in $v = 1$ and $v = 2$ vibrational states.
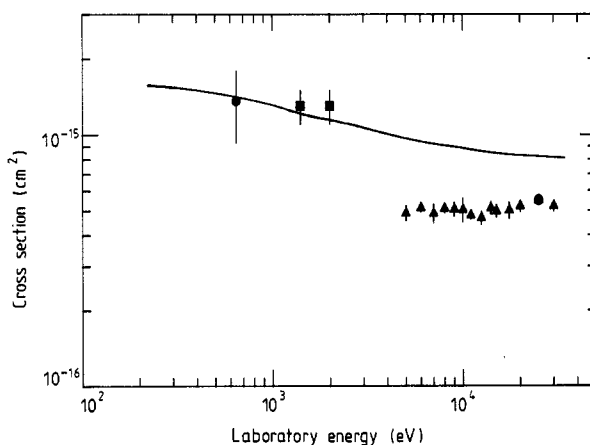

Figure 4. Cross sections for one-electron capture by 4 keV Al^{2+} in molecular hydrogen. The symbols denote: ▲, present measurements; ●, measurement of Phaneuf *et al* (1985); ■, measurement of Schrey and Huber (1981); ——, present calculations.

In figure 4 the present cross sections for Al^{2+}-H_2 collisions are compared with previous data. The single experimental value of Phaneuf *et al* (1985) at 23.9 eV amu^{-1} and two values measured by Schrey and Huber (1981) at 52 and 74 eV amu^{-1} are rather larger than might be expected on the basis of the weak energy dependence exhibited by our measured cross sections. The present calculations, while in reasonable accord with the previous low-energy measurements provide cross sections which are inexplicably considerably larger than the present experimental values.

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