Evidence for double-electron capture in the H^+_9-He collision

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Abstract:

The double-electron capture process in the H_9^+-He collision has been evidenced using the accelerator mass spectrometry method associated to a multi-coincidence detection. An absolute value for the cross section is measured. The double-electron capture of a hydrogen cluster ion from a helium atom can be thought as a very localized process involving the H_3^+core ion of the molecular cluster.

I Introduction

Hydrogen is by far the most abundant element in the Universe and molecular hydrogen H_2 is known to dominate in cool regions. Otherwise, as observed in the atmosphere of Jupiter H_3^+ is supposed to have an important role in the interstellar medium as an initiator of chains of chemical reactions[1]. The effect of protonation of pure hydrogen clusters $(H_2)_n$ at low temperature has been investigated by several quantum chemical calculations[2] including quantum Monte Carlo simulations[3]. It was shown that the added proton gets trapped as a very localized H_3^+ impurity in the cluster and is surrounded by stable shells of solvating H_2 molecules.

In recent years, research in cluster physics has expanded from the study of the isolated species in the gas phase to the interactions of atomic or molecular clusters with atoms, molecules and others clusters[4]. When a beam of molecular or cluster ions collides with a gas target, several competing reactions occur involving dissociation, electron capture, ionisation, etc… In particular, electron capture processes in ion-atom collisions play an important role in astrophysics, atmospheric physics and plasma physics[5]. Then, many investigators have studied electron-capture processes by protons[6] and molecular hydrogen ions[7] on various targets due to the inherent importance of this fundamental process. Double-electron capture is a particularly interesting case of a two-electron process[8]. Up to now most of the work has been done by measuring double-electron-capture cross sections of various atoms by multiply charged or singly charged ions.

In this paper we report on double-electron capture cross section measurements by hydrogen cluster ions (H_9^+) on helium atoms at intermediate velocities of 1.55 v_0 (60 keV/amu) where v_0 is the Bohr velocity. The protonated hydrogen cluster H_9^+ represents a specific system where a quantum solute is solvated by a quantum solvent; the added proton becomes trapped and a tightly localized H_3^+ core is surrounded by solvating H_2 molecules $H_3^+(H_2)_3$. Using the multi-coincidence technique for simultaneous detection (event-by-event basis) of correlated ionised and neutral fragments from $H_3^+(H_2)_3$-He collisions, allows us to investigate for the first time the occurrence of a double-electron capture process in cluster-atom collisions at an intermediate velocity (60 keV/amu) in a quantitative manner (absolute cross section).

II Experimental set-up

Mass selected hydrogen cluster ions with an energy of 60 keV/amu have been prepared in a high-energy cluster facility consisting of a cryogenic cluster jet expansion source combined with a high performance electron ioniser and a two step ion accelerator (consisting of an electrostatic field and a RFQ post-accelerator)[9]. After momentum analysis by a magnetic sector field, the mass selected high-energy projectile pulse (pulse length of ≈ 100 ms, repetition frequency of ≈1 Hz) consisting of $H_3^+(H_2)_3$ cluster ions is crossed perpendicularly by a helium target beam effusing from a cylindrical capillary tube (see Fig. 1). Prior to this, the ion beam is collimated by two apertures ensuring an angular dispersion of about ± 0.8 mrad. One meter behind this collision region the high-energy hydrogen collision products (neutral and ionised) are passing a magnetic sector field analyser. The undissociated primary $H_3^+(H_2)_3$ cluster projectile ions or the neutral and charged fragments resulting from the reactive collisions are then detected approximately 0.3 μs after the collision event with a multi-detector device consisting of an array of passivated implanted planar silicon surface-barrier detectors located at different positions at the exit of the magnetic analyser. Then, for each collision, all the fragments are detected simultaneously.

III Results and discussion

With this instrument we are able to record for each event simultaneously the number (multiplicity) of each mass-identified fragment ion resulting from the interaction (for more experimental details see[10,11,12]). In addition, for each event we can also monitor the sum of the masses of all the neutral fragments in coincidence with the detected ions. Moreover, by probing the angular distribution of these neutrals in front of the detector by using a movable aperture (0.5 mm of diameter) we find that the neutral products consist of hydrogen atoms and hydrogen molecules with no larger neutral clusters present[13]. The validity of single collision conditions has been ascertained by measurements at different He target pressures and allows also to derive absolute cross sections for the occurrence of specific reaction channels[14]. In Fig. 2a we report the spectrum corresponding to the detection of the neutral fragments produced by collisions of $H_3^+(H_2)_3$ cluster ions with helium atoms[15]. The number associated with each peak corresponds to the total number (mass number) for all the neutral fragments originating from one fragmented cluster ion $H_3^+(H_2)_3$. We observe 9 separate peaks. For example we can directly observe the one-electron capture process by the $H_3^+(H_2)_3$ cluster ions. This process leads to the neutralization of the cluster and corresponds to the events in the peak (9) as described previously in reference[16]. In this paper we deal to measure the double-electron capture cross section by the $H_3^+(H_2)_3$ on helium atom from this spectrum. The double-electron capture process corresponds to the following channel:

$$H_3^+(H_2)_3 + He \rightarrow ((H_9)^-)^* + He^{++}$$

$$\rightarrow \Sigma\mu_i H_i + H^- + He^{++}$$

where $\Sigma_i i\mu_i = 8$ and μ_i is the number of H_i fragments.

The double-electron capture process is followed by the dissociation of the excited negative cluster produced. We have detected no molecular or cluster negative ions. By using the multi-coincidence data sets we have deduced the number of events corresponding to this double-electron process from those events for which we detect an H^- ion in coincidence with neutral fragments of a total mass number equal to 8. In Figure 2 we report the spectra obtained in the $H_3^+(H_2)_3$ + He collision, for the detected neutral fragments (2a), for the detected H^- ion (2b), and for the neutral fragments detected in coincidence with an H^- ion (2c). In Figure 2c, the peak corresponding to mass 9 has disappeared since one H^- ion is detected. The double electron capture process corresponds to the events in peak 8. The other peaks in Figure 2c correspond to reaction channels where an H^- ion is produced simultaneously with at least one positive ion. In Figure 3 are reported the same spectra without He target obtained for the same number of incident clusters. There is no event corresponding to the double electron capture (peak 8 in Figure 2c) in the spectrum without He gas target. The one-electron capture events have been used to deduce from these data the absolute cross section for the double-electron capture. We have measured the branching ratio between the number of events (N_{dec}) in the neutral "coincident" peak (8) in Figure 2c and the number of events in the peak 9 (N_{oec}) in Figure 2a that corresponds to the total number of events for one-electron capture in the same data set.

$$R_{dec/oec} = N_{dec} / N_{oec}$$

R has been found to be equal $1.33\ 10^{-3}$.

The one-electron capture absolute cross section $\sigma_{oec,9}$ has been already measured in previous experiment[16] and found to be equal to $\sigma_{oec,9}$ = (4.4 ± 0.6) 10^{-17} cm^2. Therefore we deduce the double-electron capture absolute cross section as following

$$\sigma_{dec,9} = R_{dec/oec} \times \sigma_{oec,9}$$

The obtained value for the double-electron capture is then equal to (5.8 ± 1.2) 10^{-20} cm^2.

First we note that the value obtained for the double electron capture is very small. This process has been extracted among a large number of different and much more probable reactions induced by the collision. We have noticed in previous papers that the production of H^- ions is negligible compared to the production of other fragments. This result illustrates the power of the multi-coincidence techniques associated to accelerator mass spectrometry.

The measured cross section has to be compared with the results available in literature of proton impact on helium gas. Figure 4 shows reported theoretical and experimental values for the double-electron cross section in the 10-1000 keV/amu energy range obtained with incident protons[17]. We can observe that the measured value for the H^+ incident ion is two times larger than the corresponding value obtained for the H_9^+ ion at 60 keV/amu. From our previous results in reference 14 we show that the measured value for the H^+ ion $\sigma_{oec,1}$ (≅7.8 10^{-17} cm^2) is about twice as large as the corresponding value obtained for the H_9^+ ion $\sigma_{oec,9}$ for the same velocity. Thus, the branching ratio between double-electron capture and one-electron capture seems to be nearly the same for protons and for cluster ions at the same velocity.

In a previous paper[16] we showed that for hydrogen clusters the one-electron capture cross section is independent of the cluster size. The mean value of the one-electron capture cross section for clusters and H_3^+ has been found to be nearly the same. That shows that the electron capture by hydrogen clusters from a helium atom is a process involving mainly the H_3^+ core ion and confirms the localization of the charge on the H_3^+ core. We could interpret the present result on double-electron capture process in the same frame: that is a very localized process involving the H_3^+core ions of the H_9^+ cluster. An interesting result is the fact that this very localized double electron process is not prevented by the presence of three molecules around the H_3^+ core.

IV Conclusion

To our knowledge, this result is the first evidence for a double-electron capture process in H-cluster-ion atom collision. The value of the cross section measured in the intermediate relative velocity range is very small but only a factor 2 smaller than the double electron capture cross section in the H^+-He collision at the same velocity. This process may thus be non negligible in some ion/atom collisional systems[18,19] and should be taken into account in further theoretical investigations. Strong electron correlation have to be considered with regard to the number of electrons involved in such double capture processes.

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Figure captions

Figure 1: Experimental set-up

Figure 2:The spectra obtained in the $H_3^+(H_2)_3$ + He collision :

a) for the detected neutral fragments

b) for the detected H^- ion c) for the neutral fragments detected in coincidence with an H^- ion

Figure 3: The same spectra as in Fig. 2 without He target obtained for the same number of incident clusters:

a) for the detected neutral fragments

b) for the detected H^- ion c) for the neutral fragments detected in coincidence with an H^- ion

Figure 4: Theoretical and experimental values for the double-electron cross section in the 10-1000 keV/amu energy range obtained with incident protons[17]

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