4.8 DISSOCIATIVE RECOMBINATION OF D_3O^+ AND H_3O^+: ABSOLUTE CROSS SECTIONS AND BRANCHING RATIOS

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4.8.1 Introduction

Dissociative recombination (DR) is a process in which a molecular ion recombines with a free electron and subsequently dissociates into neutral fragments. This process occurs in the interstellar medium as well as in the inner coma of comets, planetary ionospheres and ionised layers of Earth's upper atmospheres. In these plasmas, polyatomic ions are produced in sequences of ion-molecule reactions and can be efficiently destroyed by dissociative recombination, which is typically very fast at low temperatures. To be able to understand why molecules are present in the observed amounts and to forecast the evolution of clouds, knowledge of branching ratios is required.

4.8.2 Experimental technique

Absolute cross sections and branching ratios for DR of D_3O^+ and H_3O^+ were measured at the heavy-ion storage ring CRYRING. The D_3O^+ ions were produced in a hot filament ion source (MINIS) from a mixture of D_2O and Ar gas. The H_3O^+ ions were produced in a cold plasma ion source (JIMIS) from H_2O vapour. After extraction from the source at 40 keV, the ions were mass selected and injected to the ring. They were then accelerated to a few MeV, 4.40 MeV in the D_3O^+ experiment and 5.05 MeV in the H_3O^+ experiment, and stored during a few seconds. This storage time allows infrared-active ions like H_3O^+ to relax vibrationally prior to the dissociative recombination measurement. This is very important, since under interstellar conditions (T=10–100 K) almost all the molecular ions are in their lowest vibrational level. In the electron cooler section, the ion beam merges with a collinear monoenergetic electron beam. These electrons are used both for phase-space cooling of the ions and as a target for dissociative recombination. The interaction energy is changed by tuning the electron energy. The neutral fragments created by DR follow a straight line after the electron cooler and are detected with an energy sensitive surface-barrier detector in the 0 degree arm of CRYRING. MCA and MCS spectra are recorded during the experiment to collect the DR events versus energy or versus time. For branching ratios measurements, a grid with a known transmission (T = 0.297) is additionally inserted in front of the detector.

4.8.3 Results

4.8.3.1 Branching Ratios

The branching ratios were determined over a collision energy range from 0 eV to 0.005 eV for D_3O^+ and at 0 eV for H_3O^+. Four dissociation channels are energetically allowed for DR of X_3O^+ (with X standing for H or D):

$$X_3O^+ + e \rightarrow \begin{cases} X_2O + X + 6.4\text{eV} & (n_\alpha) \\ OX + X_2 + 5.7\text{eV} & (n_\beta) \\ OX + X + X + 1.3\text{eV} & (n_\chi) \\ O + X_2 + X + 1.4\text{eV} & (n_\delta) \end{cases}$$

The branching ratios are constant over the studied energy range and no isotope effect has been observed. The results are:

$$\begin{aligned} n_\alpha(D_2O + D) &= 0.17 \pm 0.05 \\ n_\beta(OD + D_2) &= 0.13 \pm 0.03 \\ n_\chi(OD + D + D) &= 0.70 \pm 0.06 \\ n_\delta(O + D_2 + D) &= 0.00 \pm 0.04 \end{aligned}$$

$$\begin{aligned} n_\alpha(H_2O + H) &= 0.18 \pm 0.05 \\ n_\beta(OH + H_2) &= 0.11 \pm 0.05 \\ n_\chi(OH + H + H) &= 0.67 \pm 0.06 \\ n_\delta(O + H_2 + H) &= 0.04 \pm 0.06 \end{aligned}$$

4.8.3.2 Cross sections

Absolute cross sections have been determined from 0.001 eV to 0.25 eV center-of-mass energy for D_3O^+ and from 0.001 eV to 28 eV for H_3O^+. The cross sections are large (7.3×10^{-13} cm^2 for D_3O^+ and 3.3×10^{-12} cm^2 for H_3O^+ at 0.001 eV). At low energies, the cross-sections for D_3O^+ are E^{-1} energy dependent whereas it is slightly steeper for H_3O^+. A similar E^{-1} energy dependence was also observed by Mul *et al.* [1] with a merged electron-ion beam technique for both H_3O^+ and D_3O^+ and by Vejby-Christensen *et al.* [2] with the ASTRID storage ring in Denmark, who presented relative cross sections for H_3O^+. A resonance has been observed around 11 eV for H_3O^+. It reflects an electron capture to Rydberg states converging to an excited ionic core.

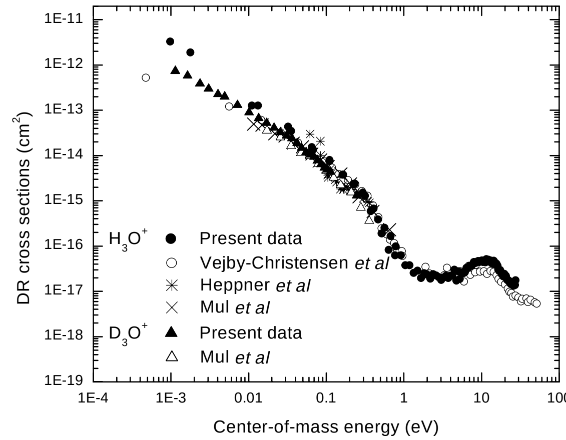

Fig 1. Effective dissociative recombination cross sections of D_3O^+ and H_3O^+ as a function of the collision energy.

A similar structure was reported by Vejby-Christensen *et al.* [2]. Our absolute measurements are in fairly good agreement with those from Mul *et al.* [1], which were first divided by 2 (Mitchell 1999, private communication) and from Heppner *et al.* [3] for H_3O^+.

4.8.4 Acknowledgements

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4.8.5 References

[1] P. M. Mul, J. Wm. McGowan, P. Defrance, and J. B. A. Mitchell, J. Phys. B **16** (1983) 3099.

[2] L.Vejby-Christensen, L.H. Andersen, O. Heber, D. Kella, H. B. Pedersen, H. T. Schmidt, and D. Zajfman, Astrophys. J. **483** (1997) 531.

[3] R. A. Heppner, F. L. Walls, W. T. Armstrong and G. H. Dunn, Phys. Rev. A **13** (1976) 1000.