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Dissociative Recombination in an Ion Storage Ring

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ABSTRACT

The astrophysically important molecular ions $DCND^+$ and D_3O^+ can dissociate upon capture of a free electron, a process known as dissociative recombination (DR). We give here a brief summary of recent experimental studies of the DR of these molecules that have been carried out at the TSR heavy ion storage ring.

1. Introduction

Dissociative recombination (DR) of molecular ions plays a key role in controlling the charge density and composition of the cold interstellar medium (ISM). Experimental DR data and reliable predictions based on a good knowledge of the underlying quantum mechanisms are required in order to understand the ISM chemical network and related processes such as star formation from molecular clouds. The required data does not only include reaction cross sections, but also the chemical composition and excitation states of the neutral products.

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DR of molecular ions involves the resonant capture of an incident electron into the potential energy surface of a doubly-excited state of the neutral molecule lying within the Franck-Condon region of the ion (Larsson & Orel 2008). Such states are usually dissociative and their asymptotic energy lies below that of the ion, reflecting the energy gained in binding the free electron. This excess energy is stabilized by dissociation into neutral, often excited fragments. Measurement of the fragment relative kinetic energies provides information on the internal states of both the parent ion and the DR products.

At the TSR heavy ion storage ring in Heidelberg, Germany, we employ a merged beams technique to study DR at low energies (Buhr et al. 2010). The ions discussed are generated in a discharge, accelerated, and injected into the storage ring. With ring pressures of $\sim 10^{-11}$ mbar, the ions can be stored for several tens of seconds. In general, ions with a dipole moment relax vibrationally – although not necessarily rotationally – to the 300 K ambient temperature. An electron beam device provides a beam of electrons with only $\sim$ 1 meV energy spread. At matching beam velocities, elastic collisions of the ions with the low-energy-spread electron beam transfer energy from the recirculating ions to the single pass electrons, thereby reducing the energy-spread of the ions (i.e., translational phase-space cooling). Thus the collision energy resolution of the experiment is defined by that of the electron beam. The electron beam velocity can be detuned from the ion velocity to investigate DR versus collision energy. The neutral DR products are not deflected by the first dipole magnet downstream of the electron beam and hit one of the dedicated DR detectors.

The studies presented here use the recently introduced Energy sensitive MUltistrip detector (EMU), which is capable of simultaneously determining both the positions and masses of all DR products. This allows the DR fragmentation channels to be distinguished on an event-by-event basis. The distribution of the kinetic energy releases (KER) is derived from the position pattern of the fragments. Knowing the basic molecular structure provides the information on the excitation level of the DR products.

2. Dissociative Recombination of D_3O^+

DR of the hydronium ion H_3O^+ is an important process for the production of H, OH and H_2O in diffuse and dense interstellar clouds. Water production is of particular interest as it is an important coolant of the ISM and this cooling contributes to the star formation rate (Neufeld et al. 1995). The branching ratios of the different DR fragmentation channels are critical parameters in modeling these cold environments. Thus, if H_2O+H is the primary channel, water will be the main reservoir of oxygen. If, on the other hand, $OH + H + H$ or $OH+H_2$ are preferred, O_2 will become the dominant oxygen-storing molecule after subsequent reactions of the OH radical.

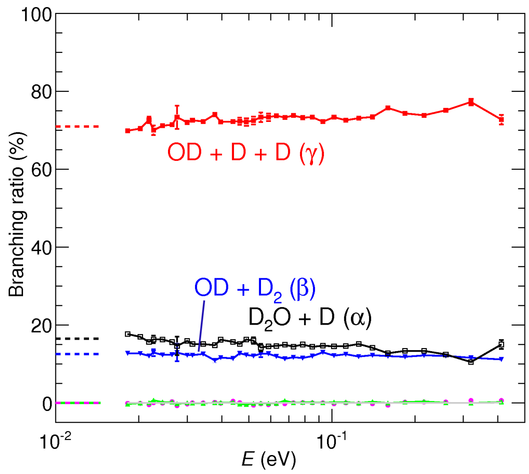

Fig. 1.— Fragmentation branching ratios for the DR of D_3O^+. The branching ratios at near-zero collision energy are marked by dashed lines. Fragmentation channels δ and ϵ are compatible with 0% over the whole energy range. The error bars represent shown only statistical errors. Additional systematic errors can reach up to 5%. Total errors at matched beam velocities are lower than 2%. See Novotný et al. (2010) for more details.

At the TSR we have investigated DR of the deuterated hydronium ion D_3O^+ employing the EMU detector. The following DR fragmentation channels can occur at low collision energies: (α) D_2O+D, (β) $OD+D_2$, (γ) $OD+D+D$, (δ) $O+D_2+D$, and (ϵ) $O+D+D+D$. With the exception for the four-body channel (ϵ), all channels are exothermic and therefore already accessible at matched beam velocities ($E \approx 0$), where the impact energies are only determined by the electron energy spread, corresponding to $\sim$ 1.0 meV. The branching ratios for the collision energy range between 18 meV and 500 meV are plotted in Fig. 1. Only the channels α, β, and γ appear to be relevant at ISM temperatures of $\sim$ 10 K. The prior experimental results of Neau et al. (2000) and Jensen et al. (2000), obtained only at matched beam velocities, are discrepant for the (α) and (γ) channels. Our work supports the results of Neau et al. and yields data for collision energies up to $\sim$ 2 orders of magnitude higher than previous work. Our novel approach of measuring branching ratios using the EMU detector results in a significantly higher precision than was previously achieved. Going from energies of 18 meV to 500 meV, only a marginal increase in the branching ratio of the γ channel and a corresponding decrease in α are observed.

3. Dissociative Recombination of $DCND^+$

Recently we have also investigated $DCND^+$, which is an isotopologue of the astrophysically important ion, $HCNH^+$. For this system, DR at near-zero collision energy has three fragmentation channels (Semaniak et al. 2001). The two isomeric channels HCN/HNC + H are expected to be the main source of HCN and HNC in the cold ISM. Observations show a source-to-source variations in the abundance ratios of these isomers which cannot be explained by astrochemical models (Hirota 1998). Thus knowledge of DR branching ratios of $HCNH^+$ forming HCN/HNC is essential for understanding the chemistry in these environments.

At the TSR we have investigated DR of $DCND^+$ at collision energies $\lesssim$ 1 meV using the EMU detector. The channels DNC + D and DCN + D cannot be distinguished through the fragment masses as they are the same for these two outgoing channels. The maximum KER for each channel is predicted to be 5.3 eV and 5.9 eV, respectively. However, the observed KER distribution for these two channels displays a broad distribution with mean value which is about 4.5 eV lower than the maximum expected values. The missing kinetic energy reflects a ro-vibrational excitation of the DCN/DNC molecular products of several eV. The majority of molecules are clearly produced with high internal excitation well above the isomerisation barrier (Bowman et al. 1993). We conclude that the DR process enables both isomers to be formed, with the final yields determined by the relaxation of the dissociated molecular products. Additional studies are needed for obtaining the branching ratios for the combined DR after the relaxation.

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REFERENCES

Bowman, J. M. et al. 1993, J. Chem. Phys., 99, 308-323

Buhr, H. et al. 2010, Phys. Rev. Lett., 105, 103202, and references therein

Hirota, T. & Yamamoto, S. & Mikami, H. & Ohishi 1998, Astrophys. J., 503, 717

Jensen, M. J. et al. 2000, Astrophys. J., 543, 764

Larsson, M., & Orel, A. E. 2008, Dissociative Recombination of Molecular Ions, Cambridge: Cambridge University Press

Neau, A. et al. 2000, J. Chem. Phys., 113, 1762

Novotný, O. et al. 2010, J. Phys. Chem. A, 114, 4870

Neufeld, D. A. & Lepp, A. & Melnick, G. J. 1995, Astrophys. J., Suppl. Ser., 100, 132

Semaniak, J. et al. 2001, Astrophys. J., Suppl. Ser., 135, 275-283
