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A NEW SETUP FOR MOLECULE FRAGMENTATION STUDIES AND THE FIRST RESULTS FROM $N^+ + H_2O$ COLLISIONS

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Abstract

A new experimental setup has been developed and installed at the right 30° beamline of the VdG-5 accelerator in Atomki. It is designed specifically for molecule fragmentation experiments [1]. Below, we describe the main features of the measuring system, and report the preliminary results of our first measurements, which were carried out by the impact of 1 MeV N^+ ions on H_2O molecules. Differential cross sections were obtained from the measured angular and energy distributions of the emitted charged fragments. Relative ionization cross sections for different degrees of ionizations were determined.

I. Introduction

Molecule fragmentation caused by energetic charged particles has attracted considerable attention in recent years [2-5]. Cross sections for the different molecule fragmentation channels are important not only from the point of view of fundamental atomic and molecular physics but in several fields of applied physics, chemistry and especially in medical applications such as radiotherapy. From an astrophysical point of view such experiments yield information about what happens in planetary atmospheres or in cometary comas during the interaction of their molecules with the solar wind [6]. In medical applications energetic charged particles such as protons or C^{q+} ions are regularly used for destroying tumorous cells [7].

In radiotherapy, cell death can be caused by direct damage of the DNA molecule or by secondary processes. In most of the direct collisions, only one side of the double helix is damaged. These so-called single strand breaks (SSB), however, can efficiently be repaired by the cell itself. Only double strand breaks (DSB) can be considered as an efficient way to damage DNA, leading to cell death. The probability of DSB, directly caused by an incident ion is typically small, therefore secondary processes have to play important role in DNA damages [8,9]. These secondary processes are in connection with the nascent free radicals originating from the fragmentation of small biomolecules by ion impact. The most important actor here is the water molecule. More than 60% of living cells is water, and they contain a large amount of carbon compounds. Accordingly, in elementary radiation safety studies, water and methane are used regularly as tissue equivalent materials for modeling living cells.

By the impact of massive charged particles (ions), radiation damage is sharply concentrated in a narrow region close to the end of the particle trajectory. This is the so called Bragg-peak region, where the linear energy transfer (LET) [10] shows a sharp maximum, and quickly drops to zero. Radiation damages, however, are not directly proportional with the linear energy transfer was found by *Montenegro et al.* [11] that the maximal fragmentation yield of the water molecule is not necessarily belongs to the top of the peak, where the LET is maximal. They have shown that the distal regions of the Bragg-peak can be even more efficient in causing fragmentation of water molecules than the Bragg maximum.

We need more experimental and theoretical work, a careful mapping of molecular collision processes for a clear understanding of the basic primary processes of radiation damage in living cells. For carbon ions, e.g., the energy region of interest falls between a few keV and ~3MeV, with charge states from 0 to 2. Fortunately, the low and medium energy accelerators in Atomki exactly suit for these regions. Accordingly, they are ideal tools for studying molecule fragmentation processes relevant for the target areas in carbon (and proton) therapy. Below, we present the results of our first molecule fragmentation experiments, which have been performed at the right 30^{O} beamline of the VdG-5 accelerator of Atomki [12] by the impact of 1 MeV N^{+} ions on water and methane gas jet targets.

II. Experimental setup

The ion beam arriving from the accelerator enters a 15^{o} electrostatic deflector. A charged state cleaned beam of the deflected ions is guided through a collimator system before entering the experimental chamber.

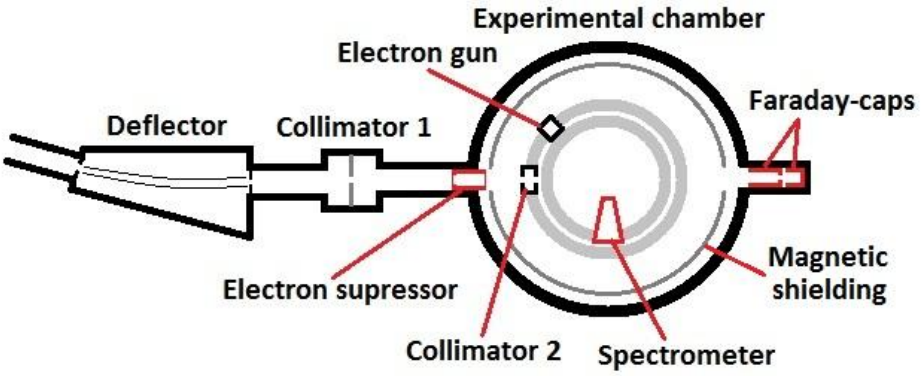

Figure 1 : The new experimental setup

A 20 mm in diameter copper tube is used as an electron suppressor electrode, mounted at the entrance of the chamber. We applied a negative voltage of -90V on it to repulse secondary electrons originating from the collimator system in the beamline. There are two, independently movable rings, centralized in the large, cylindrical (1000 x 600 mm), stainless steel, magnetically shielded experimental chamber (see Fig. 1). An energy dispersive electrostatic spectrometer was fixed on the inner ring an electron gun and a second collimator were fixed on the outer ring, which can be rotated independently from the inner ring.

After crossing the collision volume, the beam current was measured by a two-stage differential system of Faraday cups.

The electrostatic parallel plate spectrometer has been developed in Atomki, specifically for molecule fragmentation experiments [13]. Its shape allows to perform measurements in the observation angle region from 15^{o} to 165^{o} relative to the direction of the incident beam. The path length of the ions is about 10 cm

between the collision volume and the channeltron detector, for minimizing charge exchange between the background gas and the measured ionic fragments before they pass the analyser part. Electrostatic lenses, placed before the analyzer part, allow acceleration or deceleration of the studied charged particles. The energy-analyzed particles are counted by a channeltron detector. The spectrometer can be rotated around the collision volume which allows one to measure both angular and energy distributions of the charged fragments (see Fig. 2).

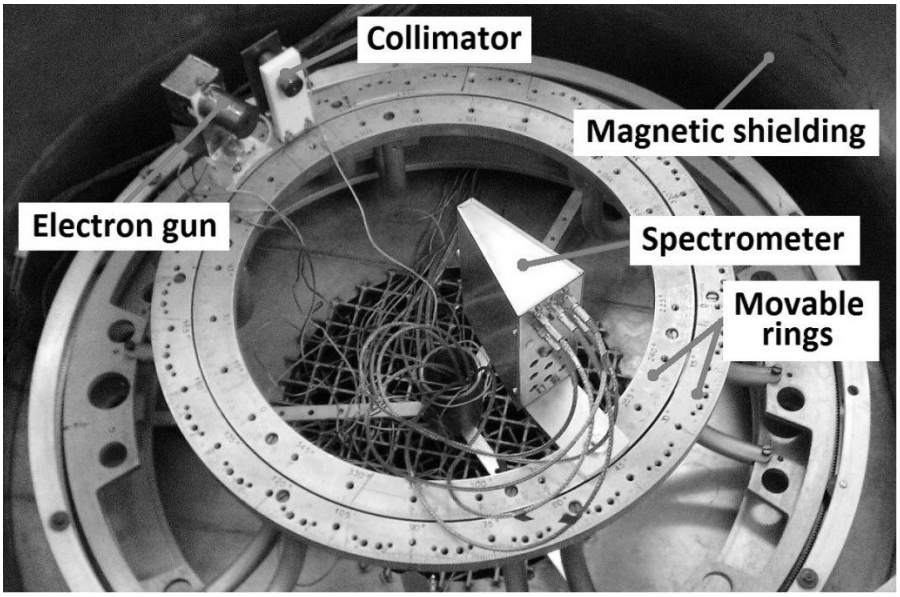

Figure 2 : Insight into the experimental chamber

The target gas is injected into the collision volume by using a special effusive gas jet target system [14]. The nozzles were designed to operate in the molecular flow range in order to provide a gas beam with small divergence and high density. Two types of nozzles were designed and tested: one with a 1 mm diameter single capillary (this was used in the experiments) and a second nozzle with a piece of a pure glass multichannel plate (MCP, fabricated for such target systems with 5 micrometer hole diameter and cca. 50 %

transparency) glued on the top of the 1 mm exit slit. The preferred flow range and target density were set by regulating the pressure in a puffer region by an automatically operated needle valve. A capillary-stricture, was applied between the puffer area and the nozzle, to ensure a convenient puffer pressure region for the regulator system. Water vapor was provided by a simple container evaporating system. Solvated contaminant gases were carefully pumped out from liquid water before the start of the measurements. During the measurements the puffer pressure was set to 20 mbar. Without target flow, the base pressure in our large vacuum chamber was about $8 \cdot 10^{-7}$ mbar.

III. Experiment

Water and methane fragmentation had been studied by several research groups [15-18], however, according to our best knowledge, data for N^+ ion projectiles have not been published yet. The charge and energy of the 1 MeV N^+ ions are similar to that belongs to the C^+ ions around the Bragg-peak, therefor the results are expected to be relevant for carbon-ion therapy. The reason of using N^+ ions instead of C^+ was technical: the lifetime of the ion source is much longer for N_2 than for carbon containing gases (CO, CO_2, CH_4, etc.). In the case of water molecule the energy of the different fragmentation channels are well-explored, so we were able to identify the channels in the $N^+ + H_2O$ collision easily.

Energy spectra were taken at the observation angles of 30^O, 60^O, 90^O, 120^O, and 150^O relative to the incident beam direction. Reproducibility of the spectra was carefully checked. The measured yields were found to be proportional to both the chamber pressure and the buffer pressure values, when the latter was varied between 10 mbar and 25 mbar.

Since the measured spectra did not show any angular dependence at the level of statistical uncertainties, average intensity values were calculated at each energy, from the data of all the angular channels. This intensity represents a fragment ion-production cross section, differential according to the energy/charge value of the fragments.

Finally, from the above cross section values, we determined the relative probabilities for the different degrees of multiple ionization.

IV. Results and discussions

In Figure 3 the normalized intensity is shown as a function of the energy in the case of water. The peaks of the spectrum were fitted by Gaussian functions in order to determine the relative probability of multiple ionization.
Double ionizations belong to the fragmentation channels marked by 2, 3, 4 and 5. These peaks fall in the 2.5 eV to 12 eV energy region as it is shown in figure 1. Triple ionizations belong only to one fragmentation channel, which is marked by number 6. The channels marked by 7 and 8 belong to 4-fold and 5-fold ionizations respectively. For more details see table 1 below.
The results shows that although the N^+ projectile ions has low charge state it can effectively cause multiple ionizations of water molecule in every fragmentation channels. According to ionization theories [19], most of the higher degree multiple ionization events may be associated with close collisions, where the projectile nucleus is only partially screened by its own electrons. The kinetic energy of the projectile is sufficiently large to provide energy for even higher degrees of ionization.

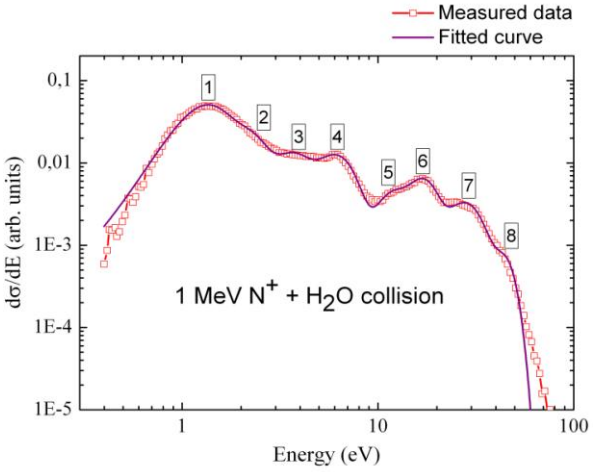

Figure 3 Relative single differential cross section for the emission of fragment ions emerging from 1 MeV N+ + H_2O collisions. Points show the experimental values, the solid line represents a fit by Gaussian functions. The numbered peaks belong to the different fragmentation channels, which are listed in Table 1. The fitted curve is also shown in the spectrum.

No. of peak	Ions	Fragm. channel	Energy (eV)	Degree of ionization	Yield
1	O^{n+}; OH^{n+}	All	1,3	All	-
2	H_2^+	$H_2^+ + O^+$	2,5	double	0,0793
3	H^+	$O + H^+ + H^+$	5	double	
4	H^+	$H^+ + OH^+$	6	double	
5	H^+	$H^+ + O^+ + H$	13	double	
6	H^+	$2H^+ + O^+$	17	triple	0,0412
7	H^+	$2H^+ + O^{2+}$	27	quadruple	0,0489
8	H^+	$2H^+ + O^{3+}$	41	quintuple	0,0100

Table 1 : Identification of the peaks and the relative yields of the different ionization degrees.

We note that peak 7 may contain both triple and quadruple ionization components [3], but we could not separate them within the present fit at this level of resolution and experimental uncertainties.

Theoretical calculation for ionization channels in the above collision system are in progress by our future collaborators.

V. Acknowledgement

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VI. References

[1] S. T. S. Kovács, P. Herczku, Z. Juhász, L. L. Horváth, B. Sulik, ATOMKI Annual Report 2010 (2011) 57.

[2] Z. Juhász, B. Sulik, F. Frémont, A. Hajaji, J.-Y. Chesnel, NIMB **267,** 326-329 (2009)

[3] P. Sobocinski, Z.D. Pešić, R. Hellhammer, N. Stolterfoht, J.-Y. Chesnel, S. Legendre, B. Sulik, NIMB **233,** 207-212 (2005)

[4] J. Tamuliene, L.G. Romanova, V.S. Vukstich, A.V. Snegursky, Chemical Physics, doi:10.1016/j.chemphys.2012.01.019

[5] U Werner, B Siegmann, H Lebius, B Huber, H.O Lutz, NIMB **205,** 639-642 (2003)

[6] H. Luna, A. L. F. de Barros, J. A. Wyer, S. W. J. Scully, J. Lecointre, P. M. Y. Garcia, G. M. Sigaud, A. C. F. Santos, V.

[7] Senthil, M. B. Shah, C. J. Latimer, E. C. Montenegro, Phys. Rev. A **75**, 042711 (2007).

[8] I. Baccarelli, F. A. Gianturco, E. Scifoni, A. V. Solov'yov, E. Surdutovich, Eur. Phys. J. D **60**, 1-10 (2010).

[9] F. Alvarado, *Ion induced radiation damage on the molecular level* (2007) p 1-7.

[10] S. Damjanovich, J. Fidy, J. Szöllősi, *Orvosi Biofizika* (2005) p 170-174.

[11] E. C. Montenegro, M. B. Shah, H. Luna, S. W. J. Scully, A. L. F. de Barros, J. A. Wyer, J. Lecointre, Phys. Rev. Lett. **99**, 213201 (2007)

[12] S.T.S. Kovács, Z. Juhász, P. Herczku, B. Sulik, Accepted ATOMKI Annual Report 2011 (2012)

[13] P. Herczku, ATOMKI Annual Report 2010 (2011) 55

[14] S. T. S. Kovacs, ATOMKI Annual Report 2010 (2011). 56.

[15] U. Werner, K. Beckord, J. Becker, H.O. Folkerts, H.O. Lutz, NIMB **98,** 385-388 (1995)

[16] N. Stolterfoht, R. Cabrera-Trujillo, R. Hellhammer, Z. Pešić, E. Deumens, Y. Öhrn, J.R. Sabin, Advances in Quantum Chemistry **52,** 149-170 (2007)

[17] J. R. Sabin, R. Cabrerra-Trujillo, N. Stolterfoht, E. Deumens, Y. Öhrn, NIMB **267,** 196-200 (2009)

[18] U Werner, B Siegmann, H Lebius, B Huber, H.O Lutz, NIMB **205,** 639-642 (2003)

[19] N Stolterfoft, R. D. DuBois, R. D. Rivarola, *Electron emission in Heavy Ion-Atom Collisions* (1997)