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Fragmentation of multiply ionized biomolecules induced by MeV heavy ions

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Synopsis Collision-induced ionization and fragmentation of gas-phase adenine molecules ($C_5N_5H_5$) have been investigated for single electron capture collisions of 1.2 MeV C^{2+} ions. Coincidence measurements between the number of ejected electrons and the time-of-flight of fragment ions allows us to determine charge states of intermediate parent ions r. Highly charged parent ions are observed up to $r \sim 6$ and the production of lighter fragment ions becomes predominant with increasing r. We found that kinetic energies of H^+ are significantly larger than those of other fragment ions and become nearly equivalent irrespective of r for r > 1.

Ion-induced fragmentation of DNA building blocks has gained increasing attention as fundamental processes in particle cancer therapy and radiation damage. In MeV heavy ion collisions, a large amount of energies are transferred from an incident ion to target electrons, leading to highly electronic excitation and multiple ionization of the target molecule. To understand the details of molecular fragmentation, we have extensively studied so far on the correlation between charged states of intermediate parent ions and fragmentation patterns for various polyatomic molecules [1, 2]. In this work, we extend measurements to an isolated biomolecular target of adenine $(C_5N_5H_5)$.

The experimental procedure is given elsewhere [1]. Correlation data between charge states of intermediate parent ions and kinetic energies (KE) of fragment ions have been obtained under a single electron capture condition of 1.2 MeV C^{2+} ions. The reaction is written as

 $C^{2+} + C_5 N_5 H_5 \rightarrow C^+ + C_5 N_5 H_5^{r+*} + n_e e^{-}$

where r represents the charge state of intermediate parent ions and is determined directly from the number of ejected electrons n_e : $r = n_e + 1$.

Figure 1 shows a result of total time-of-flight (TOF) mass spectrum of fragment ions. The main peaks are H^+ and CNH_2^+ . Compared with slow ion collisions [3], relative yields of smaller fragment ions are high because of a large amount of energy transfers. It should be pointed out that fragment ions such as CNH_3^+ , $CN_2H_3^+$ and $C_2N_3H_4^+$ are produced form proton migration in a molecule. Multiple ionization of up to $r \sim 6$ was observed by a direct electron counting technique [1-3]. By analyzing the coincidence data between the TOF and the number of ejected electrons, partial TOF spectra associated with a specific value of r were obtained. It is found that the intensities of lighter fragment ions increase with increasing r.



Figure 1. Total TOF spectrum of fragment ions obtained in collisions of 1.2 MeV C2+ with isolated adenine molecules.

Most of the fragment ions gain KE less than 10 eV, while H⁺ gains significantly large KE extending to several tens of eV. It indicates a possibility of further fragmentation of DNA by these "high energy" protons [4]. The KE distribution of H^+ varies greatly for different values of r. As for r = 1, it peaks at 0 eV, while for r > 1 the KE peaks at about 6 eV with a tail extending to about 40 eV. It is pointed out that KE distributions are nearly independent of r for r > 1. While the reason is not clear at present, the simple Coulomb explosion model may not hold in the present collision system.

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