Ab initio Molecular-Dynamics Study of Dissociation Mechanism of Highly Charged Molecules.

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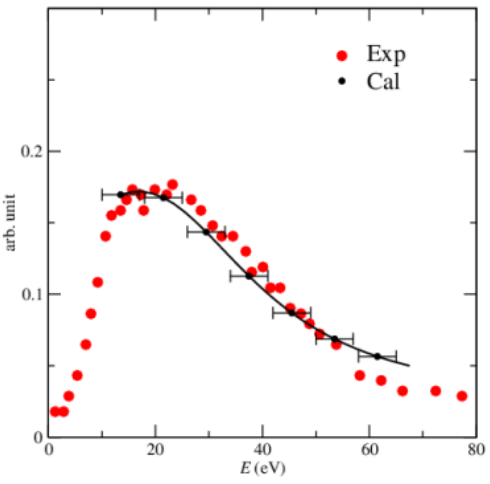
With the advent of free electron laser (FEL) such as Extreme Ultraviolet FEL and X-Ray FEL, many interesting phenomena in which atoms or molecules are highly excited due to the strong laser fields were observed. When a molecule is irradiated by intense laser, holes are proliferated through the Auger cascade and the molecule takes highly charge. The highly charged molecular ions are quite unstable due to the coulomb repulsive force and typically destroyed on a subpicosecond time scale. The fragment ions may convey information about molecular shape because they reflect geometric structure of the parent molecule just before dissociation. Recently, dissociation (Coulomb explosion) of several highly-charged aromatic molecules has been investigated by position-sensitive time-of-flight measurements. However details of the dissociation mechanism of these molecules are still unknown. Coulomb explosion mechanisms for polyatomic molecules are expected to be complicated for several reasons. First, a variety of ionic molecular fragments can be produced and there are many possible dissociation channels.

In order to clarify the atomic dissociation mechanism in the coulomb explosion of highly-charged molecules, we perform *ab initio* molecular dynamics simulation based on density functional theory. The cluster simulations for charged system were carried out using cluster boundary conditions which were imposed using the method of Martyna and Tuckerman [1]. From our simulation, it is found that molecular dissociation occurs through several stages. First, hydrogen atoms dissociate from the molecule, and then the molecule breaks into small fragments. Figure shows the kinetic energy distributions of H^+ ions of coulomb explosion of Bromo-phenol. The simulation results are good agreement with experimental results. In this study, we will discuss about the charge-state dependence of the dissociation mechanisms in coulomb explosion of several types of aromatic molecules.



[1] G. J. Martyna, M. E. Tuckerman, *J. Chem. Phys.* **110**, **(1999)**, 2810.