FRAGMENTATION DYNAMICS OF IONIZED RARE-GAS CLUSTERS: NEW ACHIEVEMENTS

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The fragmentation of rare-gas clusters Rg_n ($2 \leq n \leq 14$ and Rg = Ne, Ar and Kr) upon electron-impact ionization has been studied theoretically and compared to experiments[a]. The dynamics of these ionic clusters has been modeled by means of a trajectory surface hopping method, the Tully's Fewest Switches (TFS) method, in which all the relevant electronic states of the ions and their couplings are taken into account. A very good qualitative agreement is found for all types of clusters, concerning the extensive character of the dissociation and the tendency to form larger fragments when the parent ion size increases. For instance, no trimer fragments are found for clusters smaller than the pentamer. In addition, a very good quantitative agreement is obtained for argon clusters. On the other hand, some discrepancies are found between experiment and theory for krypton clusters: the production of monomers seems underestimated in the simulation. Theoretical results also show that the parent ion dissociation occurs within the first picoseconds, and that most of the dynamics is completed within 10 picoseconds.

Despite their success, TFS-like and adiabatic dynamics methods are based on classical mechanics and cannot reach experimental time scales, in the microsecond or millisecond range, whereas large clusters may carry on losing atoms after several nanoseconds. This issue was specifically examined on Ar_n^+ clusters (n=20 and 30): a new method that combines a TFS dynamics for the internal conversion, an electronic ground state adiabatic dynamics and phase space theory (PST) was designed and allows to reach the millisecond time scale[b].

[a] D. Bonhommeau, N. Halberstadt and U. Buck, Int. Rev. Phys. Chem. **26**, 353-390 (2007)

[b] F. Calvo, D. Bonhommeau and P. Parneix, Phys. Rev. Lett. **99**, 083401 (2007)