Charge and proton dynamics in molecules and free clusters
from atomic to nanometer scale

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2018

Popular science summary

In everyday life, we are all the time affected by processes happening on the molecular level. For instance, the weather forecast relies on models that predict the formation of clouds, a process that starts from two or more molecules sticking together in the atmosphere. Many of these processes only occur on the condition that a few atoms or molecules are held together in a certain way; much in the same way as two wheels and a frame need to be connected in the right places to make a working bicycle. To understand the processes occurring in complex molecules or collections of them, we thus need to study it when it is still intact, rather than in its separate parts.

As of now, we do not have scientific tools to follow the motion of each individual molecule and their electrons in their regular bulk environment. We can, however, extract a molecule together with its immediate neighbors, so that the result of a chemical reaction can be studied as a linked system. This collection of molecules bundled up together is called a molecular cluster. Clusters are often said to bridge the gap between a single molecule (or atom) and the macroscopic bulk liquid or solid.

In this thesis, we aim to make it possible to study complex molecules and clusters, representing simplified versions of everyday life systems. All our experiments start by shining light on clusters. The absorption of light can cause one or more electrons to leave the molecule or cluster. An electron carries a negative charge, and thus leaves the molecule or cluster positively charged, i.e. it has been ionized. Like charges repel, so if the molecular or cluster ion is doubly, or even triply charged, it can break into several pieces that are all singly charged. In molecules, we can remove electrons from a certain place in the molecule, and study the influence of different ionization locations on the types of pieces that are produced from it. In clusters, photoionization creates charges very close to each other, and we can observe which products are formed as a consequence of the charge spreading.

This thesis contributes to the development of research tools by the design of a new measurement instrument (spectrometer) and the development of methods to analyze the data from such instrument. The designed spectrometer can measure both electrons and ions, and specializes in measuring their velocities and the ion's mass.

We have studied a few molecules, for example ethyl trifluoroacetate and butadiene. Ethyl trifluoroacetate is a molecule with a very large difference in the energy required to ionize electrons from different carbon atoms in the molecule. In contrast, butadiene is a molecule with almost no difference in the needed energy to remove an electron from whatever carbon atom. We

show that the ionization of different atoms causes the molecule to break into different pieces and that the bonds around the ionization place are more likely broken in both molecules.

In addition, we have studied molecular clusters that contain ammonia and water molecules. The combination of water and ammonia is important in cloud formation in the atmosphere. We show that the molecular clusters can break up in smaller cluster fragments, or can remain intact, depending on how big they are. Ammonia and water are important in explaining the different aspects of the cluster's behavior: ammonia always carries the charges, and water seems essential in keeping the cluster intact.

Our future research will focus on molecules and clusters that become more and more similar to the ones found in the real world, for example combinations of three or more types of molecules that are thought to be important in the formation of clouds.