COST P9 RADAM Action

Short visit : 20/10/07-16/11/07
Applicant : Dr. Marta Dominika Łabuda,
Institution : Department of Theoretical Physics and Quantum Informatics,
Gdańsk University of Technology, Poland;
Host: Prof. Leticia González,
Institution: Institute for Physical Chemistry; Friedrich-Schiller-Universität Jena, Germany;

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The purpose of the Short Time Scientific Mission (STSM) visit was to start a scientific collaboration between two groups involved in the Radiation Damage in Biomolecular Systems network: the Institute for Physical Chemistry (Friedrich-Schiller-Universität Jena, Germany; and the Department of Theoretical Physics and Quantum Informatics (Gdańsk University of Technology, Poland).

The group of Prof. Leticia González (host institution) has renowned experience in the theoretical treatment of different photochemical reactions by *ab initio* quantum calculations, as well as in the field of reaction dynamics, using wave packet simulations. One of the main interests of the group is the theoretical description of reaction mechanisms, also intended to control chemical reactions using external laser fields. This is very challenging area, especially for complex molecules including many degrees of freedom. The manipulation of the internal dynamics of a particular reaction (i.e. bond breaking or isomerization) is very important in view of the very many applications that are possible in chemistry, biochemistry, physics, medicine and even nanotechnology

As the scientific objective of this STSM we wanted to study the possibility to observe the real-time charge transfer (CT) in a molecular collision, which should take place in the time scale of attoseconds (1 *as*=10^{-18} s). Our aim is to investigate for the first time nuclear and electronic wavepackets in the *as* scale in the context of ion-biomolecule collisions at keV energies. Molecular electronics, bioinformatics and cancer research may benefit from these capabilities.

We focused our attention on the theoretical studies of charge transfer occurring during collisional processes. This requires extensive calculations, which can be carried out using the MOLPRO suite of *ab initio* programs for the potentials and couplings of relevant molecular states. Optimized geometries and potential energies have been determined at the CASSCF level of theory using gaussian basis sets of atomic orbitals.

During the visit in Jena, preliminary *as* nuclear wavepackets have been simulated, assuming that the electronic character of the system (potential energy surfaces) changes along the reaction coordinate. As a suitable example, the collision of the highly charged carbon ion C^{2+} with the RNA base Uracil in one dimension (1D) has been chosen. Note that in these or similar collisional systems, the mechanism of the underlying dynamics in the CT have never been investigated. Due to the complexity of such systems, no time-resolved dynamical study has been ever attempted. Basing on the existing molecular data (potential energy surfaces and non-adiabatic couplings) previously calculated by the applicant in her PhD work (supervision of Dr. Marie-Christine Bacchus-Montabonel from Laboratoire de Spectrométrie Ionique et Moléculaire in Lyon and Prof. Józef. E. Sienkiewicz from Gdańsk University of Technology), we have extended the existing calculations to a much higher number of states which allow defining the entrance channel and a reasonable number of exchange channels. This more complex picture is necessary to start time-resolved calculations.

To solve the time-dependent Schrödinger equation (TDSE), different propagation methods have been tested. Specifically, we have chosen the Symetrize Second Order Differentiation method and the Split Operator technique, both with a discretization in a grid, which allow to describe the system in the adiabatic and diabatic representations, respectively. A tentative grid (in r- and k-space) has been chosen according to the maximum (or close to maximum) collisional velocities associated to keV energies (in these first calculations ~7eV). The computational effort scales as the first power of the number of grid points. In a 1D model, 2048 of grid points were employed as a test calculation with a very short time step. Such test calculations are computationally accessible so that the TDSE can be solved sequentially.

We have obtained a great amount of very interesting and promising results, showing the relevance of the new theoretical approaches taken to our calculations. Once these preliminary results are polished, they will be published at international journals and presented in different international conferences. This area of research can be of high importance in the areas of chemistry, biology, physics and medicine, where highly charged carbon ions are used to cancer radiation therapy. The obtained experience and results might then be used as a basis to extend the studies to other RNA or DNA bases.

This STSM gave to Dr. Marta Łabuda the opportunity to exchange and improve the knowledge on the treatment of electronic excited states and time-resolved spectroscopy of DNA building blocks, which is part of the research fields of Prof. L. González. It is our hope that our calculations will set a new line of research and motivate novel experimental investigations on time-resolved ion-biomolecule collisions. A time-dependent insight of CT processes may open the door for future developments, in chemistry and in life sciences.

This recent collaboration with the host institution will continue in the future to investigate further real time CT processes in systems of biological interest.

We take the opportunity to thank the organisers to allow this scientific exchange between these two groups, which has planted the seeds of a fruitful cooperation that will be further extended in the near future by an eventual post-doctorate stage of Dr. Łabuda at University in Jena (i. e. within Marie-Curie Intra-European Fellowship for Career Development).

Gdańsk, 14.12.2007

Dr. Marta Łabuda

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Confirmation of the Host :