Final report for the OTKA project entitled "Dynamics of molecular collisions: Experimental and theoretical studies". No: K109440

The most important results of the project are summarized briefly with references in more details.

Molecular fragmentation processes.

1. The fragmentation leading to the formation of negatively and positively charged hydrogen ions was investigated experimentally in the 7-keV OH^+ + Ar collision system. We have found that the angular and energy distribution of both the emitted negative and positive hydrogen ions show great similarity with those characterizing the elastic scattering of protons on Ar atoms. The final charge state distributions of the emitted H ions were found to follow simple statistical laws, which do not depend on the hardness of the collision. For small scattering angles (3°-30°), which correspond to soft collisions, (the centers do not get into a close encounter) one would expect a diminishing H^- formation. Instead, a rather high H^- emission rate was observed, which was explained by the excitation of the OH^+ ion before the fragmentation. See Phys. Rev. A **89** 062721 (2014) for more details.

2. Negative ion productions from gas-phase water molecules under the impact of positive ions at keV energies. Emission of both H- and heavier (O^- and OH^-) anions, with a dominant yield for H^-, was observed in 6.6-keV $^{16}O^+$ + H_2O collisions. The experimental setup allowed a separate identification of anions formed by many-body collision mechanisms from those created in hard, binary collisions. Most of the anion emission with low kinetic energy were identified as many-body collision events. Model calculations show that both nucleus-nucleus interactions and electronic excitations contribute to the observed large anion emission yield. See Phys. Rev. A **91**, 060701(R) (2015).

3. Fragmentation of the H_2O molecule in collisions with energetic H^+, He^+ and N^+ projectile ions. Energy and angular distributions of the emerging fragment ions were measured by a single-stage, parallel-plate-type, electrostatic spectrometer in a standard, crossed-beam experiment. The fragment ion emissions were found to be isotropic for all the studied projectiles. The observed differences in the fragmentation cross sections were explained by the different perturbation strengths of the bombarding particles. Cross sections for multiple ionization leading to dissociation were also deduced. The maximum observed degree of ionization was found to be q_{max} = 3, 4, and 5 for H^+, He^+ and N^+ impact, respectively. See Phys. Rev. A **96**, 032704 (2017).

4. Our experimental results on molecular fragmentation have also inspired us to investigate the process theoretically. For the description of the production of the negative and positive ions we have constructed a thermodynamical model, which applies the Boltzmann statistical law for the determination of the population of the electronic energy

levels in the quasimolecule formed during the collision. The model was applied for the OH^+ - Ar, O^+ - Ar and O^+ - Ar collisions. The calculated differential cross sections for the H^+ and H^- ion and electron emission were found in close agreement with the experimental ones. See Phys. Rev. A **94**, 022707 (2016).

Ion induced electronic transitions in molecules

We have generalized the classical Monte Carlo and quantum mechanical distorted wave methods, introduced originally for atomic collisions, for describing multicenter molecular collisions.

1. In Phys Rev. A **92,** 062704 (2015), we demonstrated the applicability of the classical trajectory Monte Carlo (CTMC) method for the treatment of the ion-induced ionization of large molecules. The treatment, within the framework the independent-particle model, is reduced to the three-body collision where the full dynamics is considered. The molecule core is described by a multicenter potential built from screened atomic potentials. The 1 MeV proton - uracil collision was considered, and our results were compared with experimental data and the predictions of quantum mechanical theories. For CTMC we found a better agreement with the experiment than for the quantum mechanical descriptions.

2. In J. Phys. B **49**, 185203 (2016) the above study was extended to the impacts of heavy ions. Reasonable agreements with the experiments were obtained for the total cross sections. However, although considerable improvements was achieved in the description of the differential cross sections, in certain ranges of the electron energy and emission angle serious discrepancies are still remained between the theory and the experiments.

3. In Phys. Rev. A **93**, 032704 (2016) the generalized continuum distorted wave with eikonal initial state (CDW-EIS) method was applied for accounting single and multiple electron removal processes (capture and ionization) in proton + H_2O collisions. Dissociation and fragmentation cross sections for the H_2O^{q+} (q=1-3) ions have also been evaluated. The results were compared with experimental and other theoretical data in a broad range of the impact energy. In general, the calculated cross sections and fragmentation yields show good agreement with the experiments.

4. In one of our joint experimental and theoretical studies electron emissions from H_2O and CH_4 molecular targets under the impacts of energetic H^+, He^+, N^+ projectile ions were investigated. Absolute double-differential electron-ejection cross sections (DDCS) were measured. Electron emissions from the projectile ions or from the target molecules and contributions of the multiple electron scattering mechanisms have also been analyzed. The DDCS data were compared with the results of the above-mentioned CTMC and CDW-EIS calculations. It was found that these methods show reasonable agreements with the measurement. See Phys. Rev. A **94**, 012704 (2016).

In our other studies, we applied the CDW-EIS and CTMC models and other quantum mechanical descriptions mostly for those molecular collision systems where fully or highly differential cross sections are available in order to check the validity of the models.

5. Ionization and electron capture reactions in diatomic molecules by impact of fast ions are processes that show large sensitivity to the degree of the coherence of the projectile beam because of the two-center scattering. In the pioneering experiment carried out for ionization of the H_2 molecule by 75 keV protons M. Schulz and his group (Missouri University, Rolla) controlled the coherence properties of the ion beam, and thereby demonstrated the effect of the projectile coherence on the angular distribution of the scattered protons. In our study presented in Phys. Rev. A **93** 012702 (2016) the effect of the loss of the projectile coherence was theoretically investigated for the 75 keV proton + H_2 collisions. Our results support the assessment that the experimentally observed broadening of the angular distribution of the inelastically scattered projectiles with decreasing projectile coherence is an inherent manifestation of the wave-packet scattering and not an experimental artifact.

6. Participating in a kinematically complete experiment, where the coherence properties of the projectile beam were controlled, we obtained new, valuable information about the role of the post-collision interaction (PCI). PCI takes place between the scattered projectile and the electron ejected into the continuum. It was found that the two-center interference structure is less pronounced under those kinematical conditions that favor the role of PCI. See J. Phys. B **48,** 175204 (2015) .

7. Single ionization and electron capture mechanisms were investigated in proton + H_2 collisions using the CDW-EIS method. In this work the accuracy of the frequently used two-effective center approximation was also discussed. For ionization, our results obtained for the various differential cross sections are in better agreement with the experimental data than those of other descriptions, especially for large momentum transfer values. For the double differential cross sections, the experimental data are reproduced quite well for both coherent and incoherent proton beam. For the charge transfer reaction, our results for the projectile angular distributions are in good agreement with the experimental data measured for coherent projectile beam. In the case of incoherent projectile, reasonable agreements with the measurements were obtained only with coherence parameters very different from those reported in the experiments. See J. Phys. B **50,** 035201 (2017); J. Phys. B **49,** 035204 (2016); J. Phys. B **51,** 035201 (2018) and The European Journal D **71**, 290 (2017).

8. Charge transfer reactions were investigated in collisions of 1-25 keV helium ions with nitric oxide. Applying the semiclassical and coupled-channel methods, total electron transfer cross sections were evaluated and compared with available experimental data. The charge transfer reaction was found to be highly anisotropic. This feature, driven by the presence of short range crossings, is characteristic to the singly charged ion +

molecule collision systems. Charge transfer processes induced by multiply charged projectile ions show a different behavior. The evaluated cross sections revealed the best agreement with the available experimental data for those collision geometries where the molecular axis is directed perpendicular to the axis of the incoming beam. See The European Journal D **68**, 167 (2014).

9. The mechanism of the electron capture was studied in the $He^+ + H_2$ collision system. The application of the four-body first Born, the CDW-EIS, the semi-classical quasi-molecular and the CTMC models enabled us to study the process in wide range of the impact energy. For impact energies above 40 keV reasonable agreements of the total cross sections have been found between the present theoretical results and the experiments. According to an analysis made in the first-Born approximation, at the highest impact energies the projectile angular distributions show interference patterns due to the coherent scattering of the projectile ions. The results were published in Eur. Phys. J. D (2017) **71**, 217.

Ion – atom collisions

We have also performed studies in the field of atomic collisions. These results also contribute to a better understanding of the molecular collision processes.

1. Charged-particle induced electron transitions in lithium atoms have received considerable attention recently, thanks to the development of a new apparatus called MOTReMi which is a combination of a magneto-optical trap (MOT) and a Reaction Microscope (ReMi). This apparatus made it possible to carry out highly sophisticated measurements. The lithium atom, due to its atomic energy level spacing, has received special interest. In our studies, we theoretically analyzed the available experimental results. We used single-active-electron (SAE), independent-electron (IEL) and beyond-independent- electron (IEL+) models to study the Li^{2+} + Li and O^{8+} + Li collisions at the experimentally considered impact energies in the MeV regime. Our calculations are based on the perturbative CDW-EIS approximation and the nonperturbative basis generator method. For outer-shell ionization, we found that the SAE picture is adequate for the determination of electron-energy dependent single-differential cross sections, but it has to be extended to allow for polarization effects, if one is interested in multi-differential cross sections. For inner-shell ionization, we found that (i) one has to go beyond the SAE picture and take two-electron excitation-ionization (EI) processes into account, (ii) doing this on the level of the IEL model, one cannot obtain satisfactory agreement with the data, i.e., electron correlation effects (IEL+ model) are likely play a role. See Phys. Rev. A **89,** 062702 (2014), Phys. Rev. A **90,** 062710 (2014), J. Phys.: Conf. Ser. **601,** 012010 (2015); and see Phys. Rev. A **93,** 012707 (2016) for more details.

In another study, the three-body dynamics of the ionization of the atomic hydrogen by 30-keV antiproton impact was investigated by calculation of fully differential cross sections (FDCSs) using the classical-trajectory Monte Carlo (CTMC) method. In the analysis, particular emphasis was put on the role of the nucleus-nucleus (NN) interaction played in the ionization process. By analyzing individual trajectories, it was found that the relative motion between the electron and the nuclei is coupled very weakly with that between the nuclei; consequently the two motions can be treated independently. The results of the calculations were compared with the predictions of available quantum mechanical descriptions. The conclusion of the comparison was that the classical mechanical description is able to reproduce the main features of the antiproton-induced ionization of the hydrogen atom, and thereby it is a very useful tool for a better understanding of the process. See Phys. Rev. A **90,** 022702 (2014) for more details.

9. There is a long standing debate in the interpretation of the fully differential cross sections measured for the 100 MeV/amu C^{6+} – He collision system (see, Nature **422**, (2003) 48). In one of our theoretical studies the role of coherent and incoherent character of the projectile beam was investigated. Cross sections evaluated in the first-Born approximation were convoluted with a Gaussian wave packet. Our results, in the scattering plane, did not show significant differences between the cross sections calculated for both coherent and incoherent projectile beam. However, in the azimuthal plane, the calculations assuming incoherent beam give a much better agreement with the experiment than those assuming coherent beam. See Nuclear Instruments and Methods in Physics Research B **408,** 165 (2017).