

MO 29: Collisions and Energy Transfer

Time: Friday 11:00–12:15

Location: F 107

MO 29.1 Fri 11:00 F 107

Vibrational autodetachment of sulfur hexafluoride anions

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We have measured the rate of electron emission from vibrationally highly excited SF₆[−] ions in a cryogenic ion beam trap. By following the emission rate over five orders of magnitudes and for up to 100 ms, we have found a deviation from the expected power function. While this is in part due to radiative cooling, we have observed the power function and the deviation from it to depend on the excitation conditions in the ion source, indicating the influence of the detachment of near-threshold states. We have therefore for the first time applied a statistical rate model that takes into account the microscopic detail of the emission mechanism and the ro-vibrational structure of the anion and the neutral molecule. Our results support the adiabatic electron affinity and the very low emission rate from states just over the threshold that have been predicted in recent theoretical investigations, with deviations being compatible with the expected effects of the rotational and vibrational temperatures of the ions and uncertainties of the model itself. We also present results from an experiment where the ions were re-excited with a laser after the initial decay had subsided.

MO 29.2 Fri 11:15 F 107

Translational, Rovibrational Enhancements and Alignments of H+ClF(v = 0-5, j = 0, 3, 6, 9)→HCl+F and HF+Cl at E_{rel}=0.5-20 kcal/mol

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QCT calculation of the title reactions on the ground PES of DHTSN of 1²A' has been done. Having the early barrier pattern, both PESs have been compared with the respective bent intermediate at angle 90° and 120°. Height of the barrier for H+ClF→HCl+F is ca. 5 kcal/mol lower, r_{HCl} and r_{ClF}, barrier region are larger than those of H+ClF→HF+Cl for both angles. Barrier location of H+ClF→HCl+F is farther from the "cliff" of the entrance and exit valleys. Larger intermediate bent angle for both reactions is associated with slightly higher barriers, larger r_{HCl} and r_{HF}, and smaller r_{ClF}. In other words, the contours of entrance valleys are slightly squeezed and precipitous; barrier regions are a little bit wider or looser. The transl. rovib. Motions of either ClF or HF in H+ClF→HF+Cl, at smaller impact parameter, via migratory and direct, should be rather difficult because of narrower entrance valley and smaller barrier region. Integral cross sections and alignments of HCl and HF with or without states v and j of ClF have been compared. Transl. vib. and rot. energy specific transl. enhancements of ClF have been found. Mechanisms on the respective PESs have been explained, and reasons of simultaneous transl. and vib. enhancements clarified.

MO 29.3 Fri 11:30 F 107

Electron Dynamics of Excitation Induced Interatomic Coulombic Decay in Quantum Dots

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The elementary physical process of interatomic Coulombic decay (ICD) that is recognized as an ultrafast energy transfer process between atoms and molecules induced by long range electron correlation, was shown to be also possible, when electrons are confined in general, non-infinite

binding potentials commonly used to model quantum dots (QDs). The electron bound to the left QD potential is radiatively excited to a higher, resonant state, after which the electron deexcites by transferring its energy to the neighboring QD potential well, from which the second electron is emitted via ICD.

Both, the excitation and the decay, were studied by means of electron dynamics in an array of two inverse Gaussian potential wells. The multiconfiguration time-dependent Hartree (MCTDH) method for antisymmetric electronic wave functions was utilized. Excitations with continuous and pulsed electric fields and with different radiation intensities were compared and the decay widths of the resonance was found to be in good agreement for the different approaches.

Besides being a tentative fundamental research topic, the realization of ICD in QDs may have important practical consequences for example in the design of infrared photodetectors.

MO 29.4 Fri 11:45 F 107

Coulomb Explosion Imaging of the metastable anion D₂[−]

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The negative hydrogen molecular ions H₂[−] and D₂[−] have been challenging theory for more than 50 years. Modern calculations predict these simple anions to be metastable when rotating strongly, with lifetimes ranging from microseconds to milliseconds. While the calculated wave functions of H₂[−] were verified by Coulomb Explosion Imaging (CEI) [1], a photoionization experiment of D₂[−] showed a deviation between theory and experiment [2]. To investigate this discrepancy we have performed a new CEI in 2012 with metastable D₂[−]. The D₂[−] ions are produced in a duoplasmatron ion source, accelerated to a kinetic energy of 1.92 MeV and directed at a diamond like carbon foil. Here the electrons are stripped off within 100 as and the Coulomb explosion is triggered. A 3D detector measures the velocity distribution of the fragments which yields a mirror image of the molecular wave function. A Monte Carlo Simulation of the explosion process is used to compare the experimental results to calculated wave functions.

[1] B.J. Thaden et al., PRL 107, 193003 (2011)

[2] L. Lammich et al., PRA 80, 023413 (2009)

MO 29.5 Fri 12:00 F 107

Low energy scattering of Stark-decelerated OH radicals with ortho- and para-H₂ molecules

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The crossed beam technique is an important experimental method to study molecular interactions, such as energy transfer or chemical reactions in collisions. The Stark deceleration technique yields control over the internal and external degrees of freedom of polar molecules in a molecular beam. The combination of these techniques opens up comprehensive investigations of molecular scattering processes as a function of the collision energy with a very high energy resolution.

Results on inelastic scattering of ortho- or para-H₂ with Stark-decelerated OH(X²Π_{3/2}, J=3/2, f) at collision energies between 40-150 cm^{−1} are presented. The main difference in the scattering process of ortho- and para-H₂ is seen in the relative cross sections, originating from the different multipole interactions involved. The ratio in the mixture of ortho- and para-H₂ was determined, in order to calculate cross sections of pure ortho-H₂ or pure para-H₂ colliding with OH, to do a comparison with theory.