MS 5: Cluster

Time: Tuesday 15:30–16:15 Location: RW 2

MS 5.1 Tue 15:30 RW 2

The Oxidation of Cationic Tantalum Clusters Under Multi-Collision Conditions — •Martin Tschurl, Jan Frederik Eckhard, Tsugunosuke Masubuchi, Daniel Neuwirth, and Ueli Heiz — Technical University of Munich, Chair of Physical Chemistry, Lichtenbergstraße 4, 85748 Garching, Germany

Tantalum clusters and their oxides represent prospective materials for catalysis. In particular, they offer great potential as oxidation catalysts, as e.g. in the electrochemical oxygen evolution reaction or the methane activation. In this talk it will be shown, how the oxidation properties of tantalum clusters are governed by their size. It is found that three different species are formed depending on the number of atoms and the oxidation state of the clusters. For smaller clusters fully oxidized tantalum oxides result, whereas the oxidation of larger clusters favors the formation of species with an intact metal core. For the latter a second oxidation mechanism occurs of higher oxidation states, which is assigned to species for which the O-O bond remains intact. Furthermore, it is show how these results will contribute to future studies of ion-molecule reactions.

MS 5.2 Tue 15:45 RW 2

Room-temperature methane activation by tantalum (oxide) clusters — •Jan Frederik Eckhard, Tsugunosuke Masubuchi, Daniel Neuwirth, Martin Tschurl, and Ueli Heiz — Technical University of Munich, Chair of Physical Chemistry, Lichtenbergstraße 4, 85748 Garching, Germany

The selective transformation of methane has received a lot of attention as it is one of the most important reactions that require C-H bond activation. Unraveling the mechanistics of such complex processes is facilitated in gas-phase studies. In our setup, the combination of a laser vaporization cluster source, a cryogenic ring electrode ion trap and a reflectron time-of-flight mass spectrometer is used to analyze reactions of size-selected metal clusters under multi-collision conditions. Doing so, reaction pathways as well as size-dependent apparent rate constants and activation energies are obtained. Additional information, e.g. about intrinsic cluster properties, may be revealed by employing ab-initio calculations. Tantalum was chosen as a clustering material as it is one of the few metals that, in its atomic cation form, brings about hydrogen elimination from methane. Cationic tantalum clusters and tantalum oxide clusters are also found to activate methane in consecutive dehydrogenation reactions. These new results will be presented in detail and surprising findings concerning cluster size, composition and activation energies will be discussed.

MS 5.3 Tue 16:00 RW 2

High-resolution cluster analysis by multi-reflection time-of-flight mass spectrometry — Paul Fischer[1], •Stefan Knauer[1], Gerrit Marx[1], Birgit Schabinger[1], Lutz Schweikhard[1], and Robert Wolf[2] — [1]Institut für Physik, Universität Greifswald, Felix-Hausdorff-Str. 6, 17489 Greifswald — [2]ARC Centre of Excellence for Engineered Quantum Systems, School of Physics, The University of Sydney, NSW 2006 Australia

Multi-reflection time-of-flight (MR-ToF) devices [1] have established themselves as mass separators and spectrometers with high resolving powers and fast processing times. A MR-ToF analyzer built at Greifswald was installed at ISOLTRAP for the investigation of exotic nuclei [2]. Based on this design a further setup combining MR-ToF analyzer with a laser ablation source was constructed for atomic cluster research. The developments aim to provide samples of selected clusters for further investigations, such as cluster-laser interactions or photoelectron spectroscopy. A characterization of the setup and first experimental results will be presented.

[1] H. Wollnik, M. Przewloka: Int. J. MassSpectrom. Ion Proc. 96 (1990) 267

[2] R. Wolf et. al, Phys. Rev. Lett. 110 (2013) 041101; F. Wienholtzet. al, Nature 498 (2013) 346;M. Rosenbusch et. al, Phys. Rev. Lett. 114 (2015) 202501; D. AtanasovPhys. Rev. Lett. 115 (2015) 232501