A 18: Helium Droplets and Systems (with MO)

Time: Tuesday 14:30–16:15 Location: N 6

Invited Talk A 18.1 Tue 14:30 N 6

Cluster Studies with the BerlinTrap — •Pablo Nieto, Alan Günther, David Müller, Alex Sheldrick, and Otto Dopfer — Institut für Optik und Atomare Physik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany

The first experiments using a novel tandem mass spectrometer (BerlinTrap) which combines an electrospray ion source, a quadrupole mass spectrometer, a cryogenic 22-pole ion trap (4-300 K), and an orthogonal reflectron time-of-flight mass spectrometer [1] are presented. First, the generation of small He clusters around cold metal ions (He_nM^{q+}) by successive aggregation of He atoms is presented. The attraction between a positive charge and the surrounding He leads to the formation of a shell of strongly bonded atoms around the ion often called snowball. The study of He snowballs has been carried out in the last years by photo- or electron impact ionization of the atom or molecule embedded in relatively large He droplets. This study constitutes the first bottom-up approach to the formation of He snowballs around an ion using a gentle growth scheme. The formation of some clusters with a given (magic) number of He atoms is greatly enhanced signaling a high relative stability. In the second part of the talk structure and optical absorption properties of metalated flavins in the gas phase are discussed. The present results illustrate the broad variety of atomic and molecular ionic clusters which can be studied with the BerlinTrap by mass spectrometry and spectroscopy.

[1] A. Günther, P. Nieto, D. Müller, A. Sheldrick, D. Gerlich, and O. Dopfer, J. Mol. Spectrosc., in press, DOI:10.1016/j.jms.2016.08.017

A 18.2 Tue 15:00 N 6

Libration of strongly-oriented polar molecules inside a superfluid — •Elena Redchenko and Mikhail Lemeshko — IST Austria (Institute of Science and Technology Austria), Am Campus 1, 3400 Klosterneuburg, Austria

Recently, it was shown that molecular rotation inside a superfluid, such as a helium nanodroplet or a Bose-Einstein condensate, leads to formation of a quasiparticle of a new kind – the angulon. The angulon represents a quantum rotor dressed by a field of many-particle excitations. In this work, we demonstrate that in the strong-field limit the angulon turns into the 'pendulon' – a quantum spherical librator, whose pendular motion is altered by the field of phonon excitations, and study the properties of the pendulon quasiparticle. We have shown that an electric field relaxes the selection rules on the angular momentum exchange between the molecule and the bath, which results in a series of instabilities absent in the field-free case of the angulon quasiparticle. In other words, a field renders the instabilities universal, i.e. independent on the details of the molecule-boson potential energy surface. Furthermore, a field acts as an additional knob for altering the positions of the instabilities in the molecular rotational spectrum, thereby opening the door for detailed experimental studies of redistribution of orbital angular momentum in many-particle systems.

[1] Redchenko E.S., Lemeshko M., Chemphyschem. 2016 Nov 18;17(22):3649-3654.

A 18.3 Tue 15:15 N 6

Fingerprints of the angulon quasiparticle in spectra of molecules in superfluid helium droplets — •Igor Cherepanov and Mikhail Lemeshko — Institute of Science and Technology Austria (IST Austria), Am Campus 1, 3400 Klosterneuburg, Austria

The recently developed angulon theory [1] represents a powerful tool to study interactions between a rotating molecular impurity and a macroscopically large number of helium atoms in a superfluid helium nanodroplet. The comparison of the experimental data on the renormalization of rotational constants for a wide range of different molecules and the predictions of the angulon theory [2] provides a strong evidence for angulon formation in superfluid ^{4}He.

Here we demonstrate that angulon theory is also able to explain some features observed in the ro-vibrational spectra of the ν_3 band of the methyl radical [3] and the ammonia molecule [4] solvated in superfluid ^{4}He. We found that one of the largest anisotropic terms in the $CH_3(NH_3)$-He potential expansion induces a transfer of one quantum of angular momentum from the molecule to the many-body bath. As a consequence, the spectral line corresponding to the $^RR_1(1)$ transition becomes significantly broadened and suppressed compared to the gas phase. This amounts to an experimental confirmation of the angulon instabilities, recently predicted in Ref. [1].

[1] R. Schmidt, M. Lemeshko, Phys. Rev. Lett. **114**, 203001 (2015)
[2] M. Lemeshko, arXiv:1610.04908 (2016)
[3] A. Morrison et al., J. Phys. Chem. A **117**, 11640 (2013)
[4] M. Slipchenko, A. Vilesov, Chem. Phys. Lett. **412**, 176 (2005)

A 18.4 Tue 15:30 N 6

Gold doped helium nanodroplets: Electronic spectroscopy from atoms to nanoparticles — •Florian Lackner, Maximilian Lasserus, Roman Messner, Martin Schnedlitz, Alexander Volk, Philipp Thaler, and Wolfgang E. Ernst — Institute of Experimental Physics, TU Graz

The isolation of nanoparticles in helium nanodroplets offers low temperature confinement for a dopand size regime ranging from single atoms and molecules to large clusters and elongated wire-structures. Excitation spectra of gold atoms, residing inside the droplets, appear strongly blue shifted due to the repulsive Pauli interaction of excited electrons with the He environment. Consequently, for the $5d^{10}6p\ ^2P^o_{1/2} \leftarrow 5d^{10}6s\ ^2S_{1/2}$ transition in Au-He_N we observe a broad structure extending more than 800 cm^{-1} to the blue of the bare atom line located at 37,359 cm^{-1}. This feature is superimposed by a peak around 37,950 cm^{-1} attributed to atoms that have relaxed into the $5d^96s^2\ ^2D$ manifold, a pathway enabled by the droplet environment and forbidden in the free atom. Absorption spectra of larger nanoparticles can be recorded ex-situ after deposition on fused silica substrates. First results on Au nanoparticles prepared by helium nanodroplets show a characteristic surface plasmon resonance peaking around 530 nm. We expect that the formidable doping capabilities provided by helium nanodroplets will allow for the study of the electronic structure of a large variety of mono- and bi-metallic nanoparticles ranging from single atoms and molecules to large nanoparticles.

A 18.5 Tue 15:45 N 6

Far-Infrared Spectroscopy of Molecules and Aggregates in Helium Nanodroplets at FELIX — •Gerhard Schwaab[1], Raffael Schwan[1], Devendra Mani[1], Nitish Pal[1], Britta Redlich[2], Lex van der Meer[2], and Martina Havenith[1] — [1]Physikalische Chemie 2, Ruhr-Universität Bochum — [2]FELIX Laboratory, Radboud University Nijmegen

Helium nanodroplets provide the opportunity to study molecules and molecular aggregates at temperatures of 0.37 K in a suprafluid matrix. Recently we have moved one of our helium droplet machines to the free electron laser facility FELIX in Nijmegen. First test molecules included SF_6, water, water clusters and water-HCl clusters.

The high output energy and broad frequency coverage of the FELIX I and FELIX II beamlines allows access to the low frequency ($\nu > 100 cm^{-1}$) far-infrared range in spite of the low energy per photon. Esecially the spectrum of small water clusters in this region is astonishingly rich. An overview of the experimental setup and the results of our latest micro-solvation studies will be given.

A 18.6 Tue 16:00 N 6

Infrared spectroscopy of HCl dissociation at 0.37K using free electron lasers. — •Devendra Mani[1], Raffael Schwan[1], Theo Fischer[1], Arghya Dey[1,2], Matin Kaufmann[1], Britta Redlich[2], Lex van der Meer[2], Gerhard Schwaab[1], and Martina Havenith[1] — [1]Lehrstuhl für Physikalische Chemie II, Ruhr Universität Bochum, Germany. — [2]Institute for Molecules and Materials (IMM), Radboud University Nijmegen, Nijmegen, Netherlands

Dissociation of HCl in presence of few water molecules has recently been studied theoretically as well as spectroscopically. [1,2] These studies suggest that 4 water molecules are sufficient to dissociate one HCl molecule, forming a solvent separated $H_3O^+(H_2O)_3Cl^-$ dissociated cluster. Until now, attempts to observe this dissociated species had been focused on the O-H and H-Cl stretch regions. However, the results were obscured by the presence of vibrational bands of different $(HCl)_m - (H_2O)_n$ undissociated clusters in this spectral range. From the recent theoretical calculations [3], the umbrella type motion of the H_3O^+ moiety (prediction: broad band, 1300-1360 cm^{-1}) appears to be a fingerprint signature for the dissociated cluster. We have studied this dissociation reaction in helium droplets, in the frequency range

of 900-1700 cm^{-1}, using free electron lasers at FELIX. A weak broad band, spanning from 1000 to 1450 cm^{-1}, could be observed on specific mass channels. The results will be discussed in detail in the talk.

References: 1) H. Forbert et al., J. Am. Chem. Soc., **133**, 4062 (2011). 2) A. Gutberlet et al., Science, **324**, 1545 (2009).3) J. M. Bowman et al., Phys. Chem. Chem. Phys., **17**, 6222 (2015).