FEMTOlab, Aarhus University, presents:

Mini Symposium 2015
on
Molecules in Helium Nanodroplets and Cold Molecules

Thursday 1st October 2015, 13:15-17:00
Department of Physics and Astronomy, Room 1520-732, Ny Munkegade 120, 8000 Aarhus C

Everybody is very welcome – also to attend just a single selected talk of your favourite pick.
Abstracts are given below.

13:15-14:05: **Probing the critical Landau velocity in finite-size helium systems**
Marcel Drabbels, Laboratoire de Chimie Physique Moléculaire, Ecole polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland

14:05-14:55: **Femtosecond dynamics of doped helium droplets: pump probe imaging spectroscopy and electronic wave packets**
Frank Stienkemeier, Physikalisches Institut, Universität Freiburg, Germany

14:55-15:20: Coffee and cake

15:20-15:55: **Laser-induced alignment of molecule-He complexes**
Anders A. Søndergaard, Department of Physics and Astronomy, Aarhus University, Denmark

15:55-16:20: **Laser-induced alignment of molecules in He droplets**
Benjamin Shepperson, Department of Chemistry, Aarhus University, Denmark

16:20-17:00: **Rotational Cooling of Coulomb-Crystallized Molecular Ions by a Helium Buffer Gas in a linear rf quadrupole trap**
Michael Drewsen, Department of Physics and Astronomy, Aarhus University, Denmark

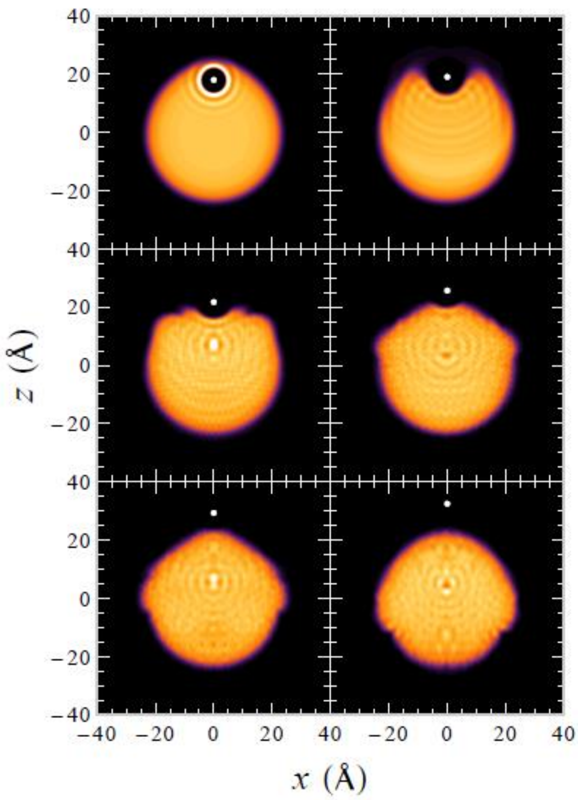
Host: Henrik Stapelfeldt

Probing the critical Landau velocity in finite-size helium systems

Marcel Drabbels

Laboratoire de Chimie Physique Moléculaire, Ecole polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

The best-known property of superfluid helium is the vanishing viscosity that objects experience while moving through the liquid with speeds below the so-called critical Landau velocity. This critical velocity is generally considered to be a macroscopic property since it is related to the collective excitations of the helium atoms in the liquid. Here I will report experiments that show the existence of a critical Landau velocity in nanometer scale, finite-size helium systems. The experiments use a direct approach to probe the superfluid character of helium nanodroplets by determining the vanishing viscosity embedded atoms and molecules experience as they are being accelerated out of the droplets following optical excitation. By recording the speed distributions of the ejected species using imaging techniques we find that a critical Landau velocity exists in these systems, even for nanodroplets consisting of less than a thousand helium atoms. We furthermore find that the creation of roton pairs is at the origin of this critical velocity, and that the mean speed and the width of the speed distributions are in quantitative agreement with predictions based on the dispersion curve of superfluid helium. The experimental conclusions find further support in theoretical simulations based on a time-dependent density functional description of the helium bath.



Evolution of $AgHe_{1000}$ following excitation of the Ag atom to the 5p $^2P_{1/2}$ state.

Femtosecond dynamics of doped helium droplets: pump probe imaging spectroscopy and electronic wavepackets

Frank Stienkemeier

Physikalisches Institut, Universität Freiburg, Germany

Dynamics of helium droplet isolated species are probed by means of femtosecond laser techniques. On the one hand, nuclear dynamics of rubidium (Rb) atoms attached to helium nanodroplets is characterized by ion and electron imaging techniques. E.g., the desorption of Rb from the droplet surface is initiated by a femtosecond pump pulse. Upon a delayed ionizing probe pulse, velocity map ion and electron images as well as time of flight traces are recorded. From the resulting observables desorption times $t \leq 10$ps are derived, accompanied by formation of $Rb^+He_{n=1,2}$ complexes. The observed desorption process is convoluted with the submersion of Rb ions into the droplet interior for time delays $t \leq 2$ps, which we infer by detecting high cluster masses with an optimized TOF mass detection unit. On the other hand electronic wavepackets have been probed implementing a phase modulation scheme into the femtosecond pump-probe setup. Applying this technique to Rb-doped helium nanodroplets allows to detect the formation of RbHe exciplex molecules vibrationally resolved. Spectral resolution as well as sensitivity comes out to be improved by orders of magnitudes when compared to standard pump-probe measurements. Furthermore, phase cycling allows us to efficiently isolate multiple coherences. Even at very dilute targets, fifth-order coherences have been measured and are interpreted in terms of collective ionization processes.

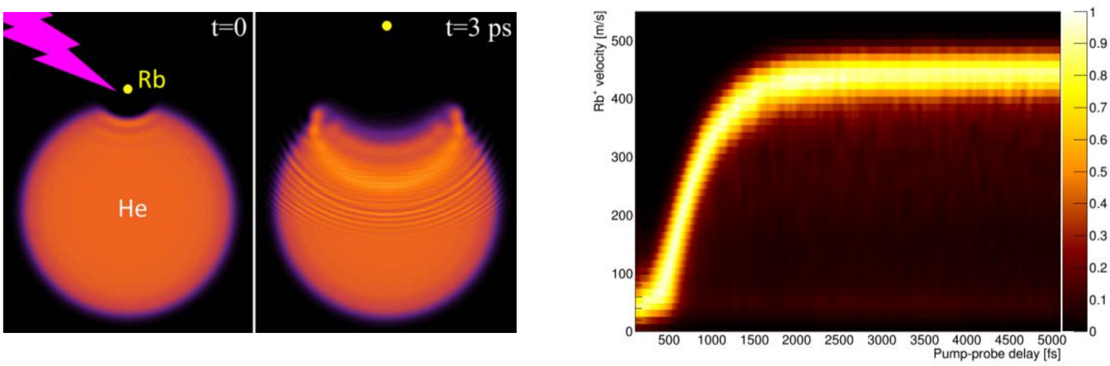


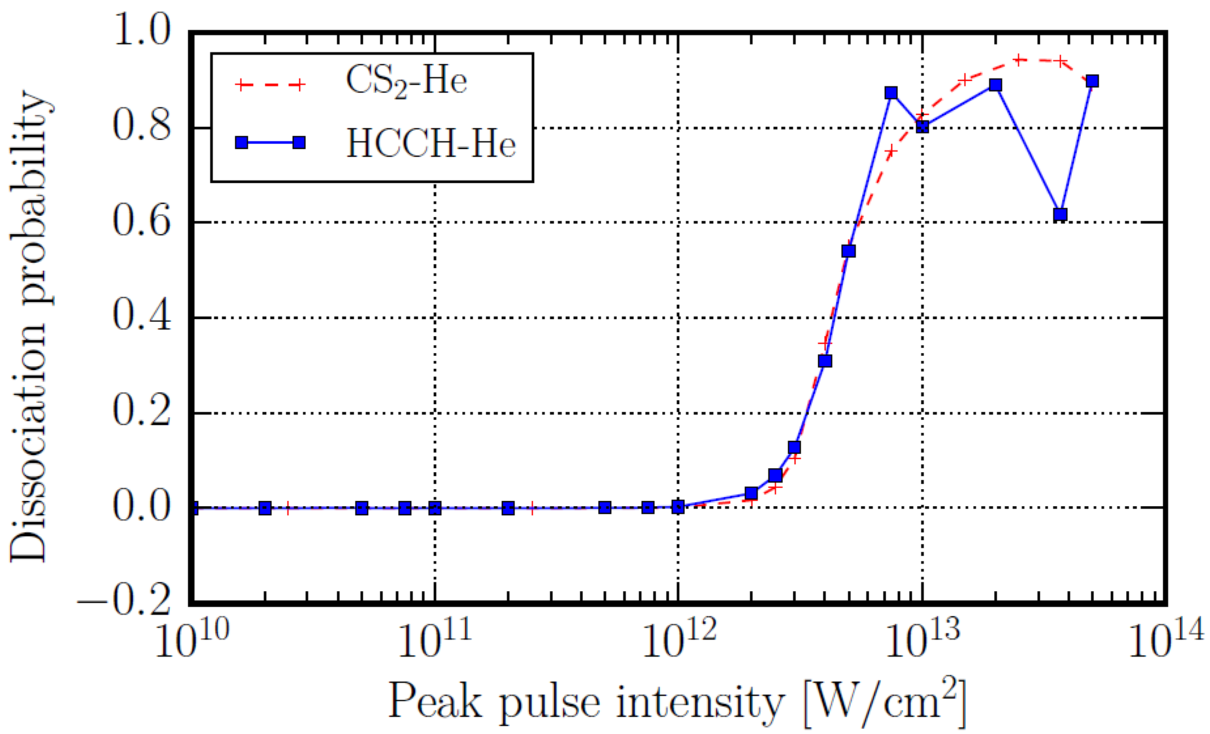
Figure 1: Left: Simulated initial geometry and time evolution of a photoexcited Rb atom attached to the droplet surface. Right: Experimental speed distribution of desorbing Rb^+ ions at different pump probe delays for laser excitation to the droplet perturbed Rb 6P state.

Laser-induced alignment of molecule-He complexes

Anders Aspegren Søndergaard

Department of Physics and Astronomy, Aarhus University, Denmark

The polarizability interaction between a molecule-He van der Waals complex and a moderately intense laser pulse is studied numerically. The molecules CS_2 and C_2H_2 are investigated and the laser pulse has a duration of 330 fs which is much shorter than the natural rotational time of the two molecular species. For high intensities of the laser pulse the complex is dissociated rapidly and the molecule undergoes alignment dynamics essentially similar to an isolated molecule. At low intensities the complex stays intact and exhibits an alignment dynamics that differs significantly from that of the isolated molecules. The results are rationalized and their relation to recent experiments is discussed. Possible implications for laser-induced alignment of molecules in Helium nanodroplets will be mentioned.



Dissociation probability for the ground state of CS_2-He and HCCH-He as function of peak pulse intensity for a 300 fs Gaussian pulse

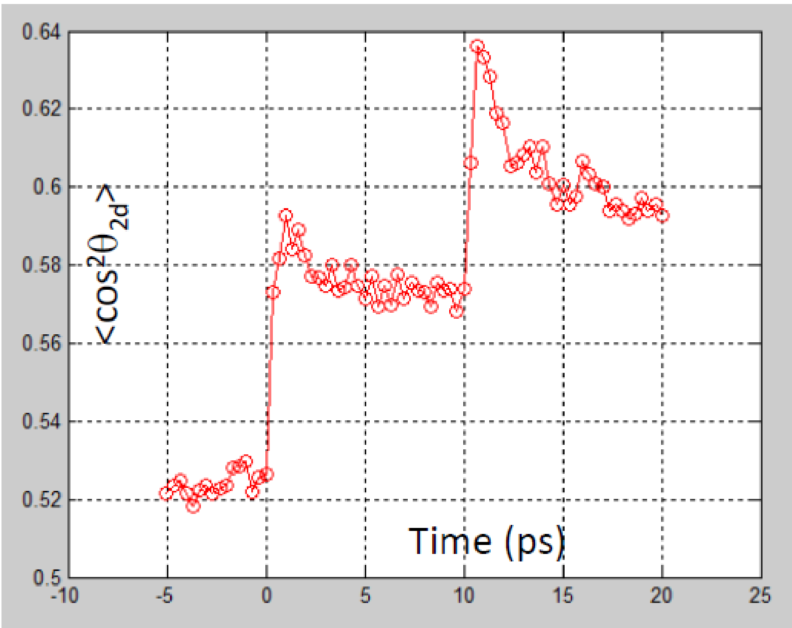
Laser-induced alignment of molecules in He droplets

Benjamin Shepperson

Department of Chemistry, Aarhus University, Denmark

Experimental studies of alignment of molecules in Helium nanodroplets induced by nonresonant laser pulses with a duration of a few hundred femtosend is discussed. Results on CS_2 molecules recorded on the first generation droplet machine from Femtolab will be presented. These results show an initial rotational dynamics almost as fast as for isolated molecules but a much slower decay of the prompt alignment – similar to previous studies on methyliodide and iodobenzene.

Preliminary test results obtained on a newly designed helium droplet machine will be presented and prospects for studies on light species like OCS, CO_2 and C_2H_2 are discussed.



The degree of alignment of CS2 molecules inside a He droplet when irradiated by two 300 fs long laser pulses – the first sent at time = 0 ps and the second at time = 10 ps

Rotational Cooling of Coulomb-Crystallized Molecular Ions
by a Helium Buffer Gas in a linear rf quadrupole trap

Michael Drewsen

Department of Physics and Astronomy, Aarhus University, Denmark

In this talk, I will discuss recent experimental results on helium buffer-gas cooling of the rotational degrees of freedom of MgH^+ molecular ions, which are trapped and sympathetically crystallized in a linear radio-frequency quadrupole trap. With helium collision rates of only ~10 s^{-1}, i.e. four to five orders of magnitude lower than in usual buffer gas cooling settings, we have cooled a single molecular ion to an unprecedented measured low rotational temperature of $7.5^{+0.9}_{-0.7}$ K. In addition, by only varying the shape and/or the number of atomic and molecular ions in larger Coulomb crystals, we have tuned the effective rotational temperature from ~7 K up to ~60 K by changing the micromotion energy. The very low helium collision rate may potentially even allow for sympathetic sideband cooling of single molecular ions, and eventually make quantum-logic spectroscopy of buffer gas cooled molecular ions feasible. Furthermore, application of the presented cooling scheme to complex molecular ions should have the potential of single or few-state manipulations of individual molecules of biochemical interest. This latter perspective can hopefully be exploited to disentangle various processes happening in complex molecules, like light harvesting complexes.