QFC2019 Overview (“wrap up”)

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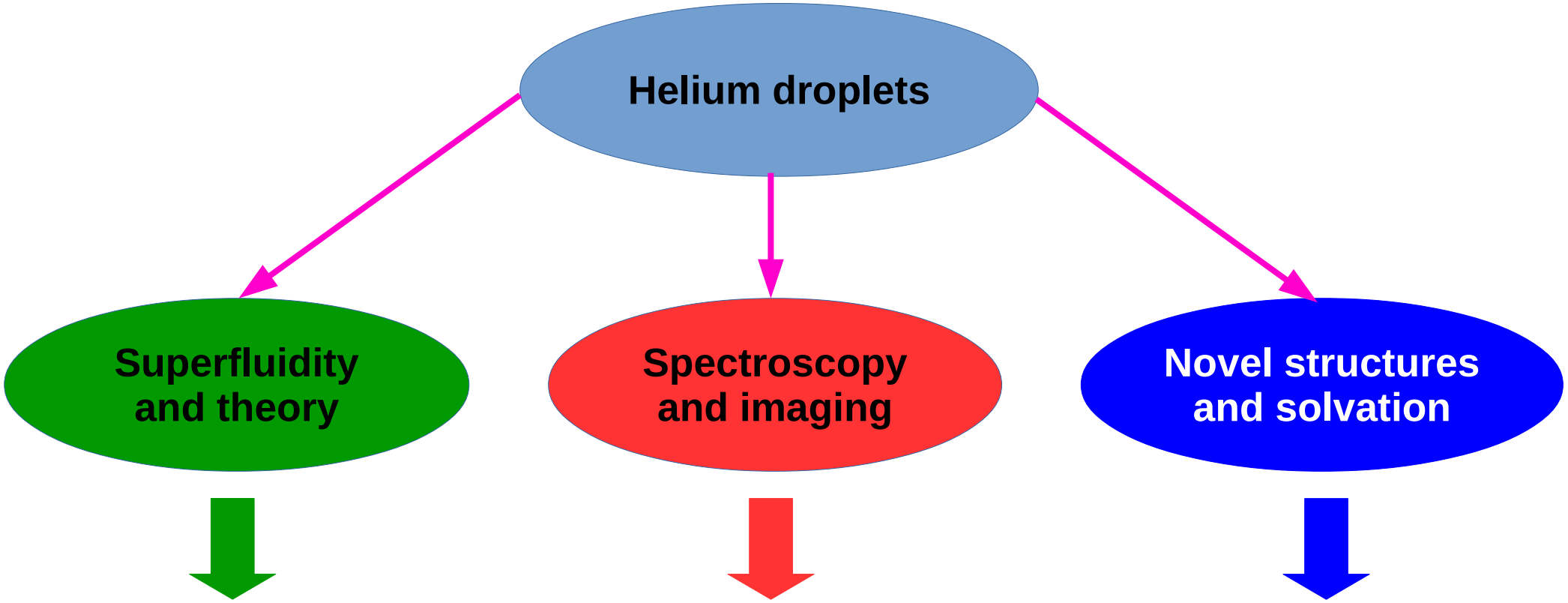
Helium droplet research “family tree”



- Finite-size effects
- Translational vs. rotational superfluidity
- Quantized vorticity
- Superfluid dynamics
- Confinement effects
- ^{3}He/^{4}He mixtures

- “Clean” solvation of atoms & molecules!
- Low temperature and weak interaction
- High-Res. spectroscopy
- Molecular alignment
- Internal excitations

- Thin nanowires at low temperatures
- Low temperature chemistry (complexes)
- Solvent-bound species (“quantum gels” etc.)
- Soft landing

Superfluidity

Quasiparticle approach to far-from-equilibrium dynamics of molecules in helium nanodroplets (Mikhail Lemeshko):

- Angulons provide a general framework to study angular momentum dynamics in quantum many-body systems
- Shown to work for rotation of molecules in (large) superfluid helium droplets
- Application: modeling of the dynamics following molecular alignment in helium droplets

Superfluidity & theory

Can 0.4 K He induce electronic relaxation? The case of Rb and Ba+ @ superfluid He droplets (Nadine Halberstadt et al.):

- Non-adiabatic crossings between electronic states (answer: yes)
- Employed both He-TDDFT and quantum molecular dynamics with non-adiabatic transitions
- He-TDDFT can model atomic scale dynamics in helium droplets very accurately

Superfluidity & theory

Superfluid helium nanodroplets: The many impurities, many vortices cornucopia (Manuel Barranco et al.):

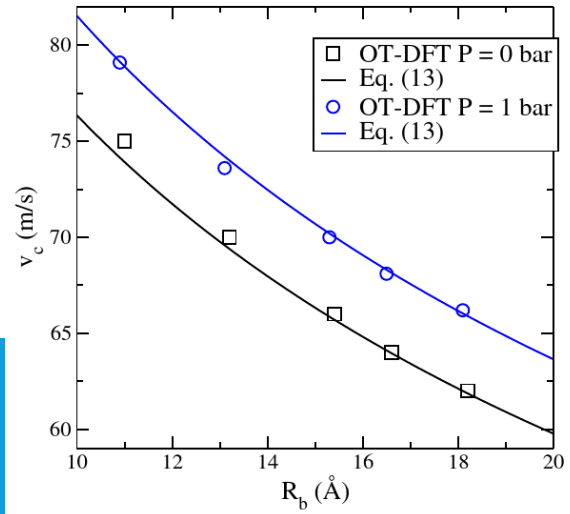
- He-TDDFT of vorticity in helium droplets and capture of impurities (Ar, Xe) as pertaining to experimental visualization of vorticity
- Modeling of superfluid droplet shapes
- “Educational speech” on vorticity in helium: vortices in superfluid helium are *irrotational* (curl is zero). Classical vorticity has non-zero curl and real flow. *Very much appreciated speech!*

Superfluidity & theory

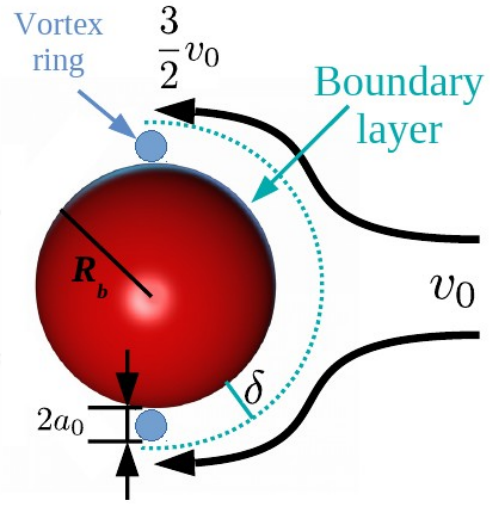
General question on Landau critical velocity:

- Landau critical velocity is related to creation of rotons
- Feynman critical velocity is related to creation of vortices
- At least to pressures up to 10 bar, the dissipation mechanism is due to Feynman. Above this pressure, Landau process may appear (?)

General question on size dependence of the critical velocity:



Kinetic energy around the bubble vs. vortex ring creation energy. Critical velocity on macroscopic scales is just few cm/s

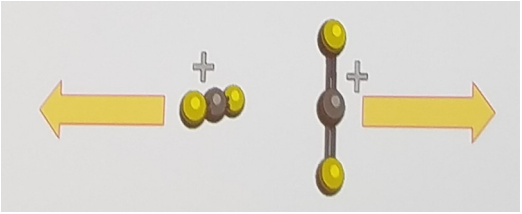


Phys. Rev. B 98, 094520 (2018)

Spectroscopy & imaging

Imaging controlled complex in helium droplets with Coulomb explosion (Adam Chatterley *et al.*):

- Determine structure and in the future dynamics of weakly bound complexes
- Cooling by helium droplets reduces the number of possible conformers – not possible in practice without this!
- Long-pulse laser alignment of molecules with subsequent ionization leading to Coulomb explosion:



Spectroscopy & imaging

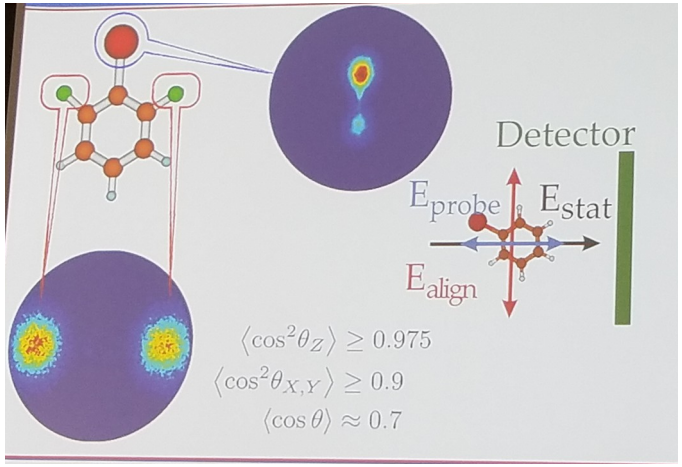
Two-dimensional coherent spectroscopy of doped helium nanodroplets (Frank Stienkemeier *et al.*):

- 2-D femtosecond spectroscopy (4-wave mixing) that yields unprecedented spectral-temporal resolution
- Rb_2: Vibrational wavepacket dynamics example
- Rb_3: Desorption dynamics from helium droplet surface
- Future application: Dynamics of charge transfer complexes

Spectroscopy & imaging

Controlled molecules and nanoparticles (Jochen Küpper):

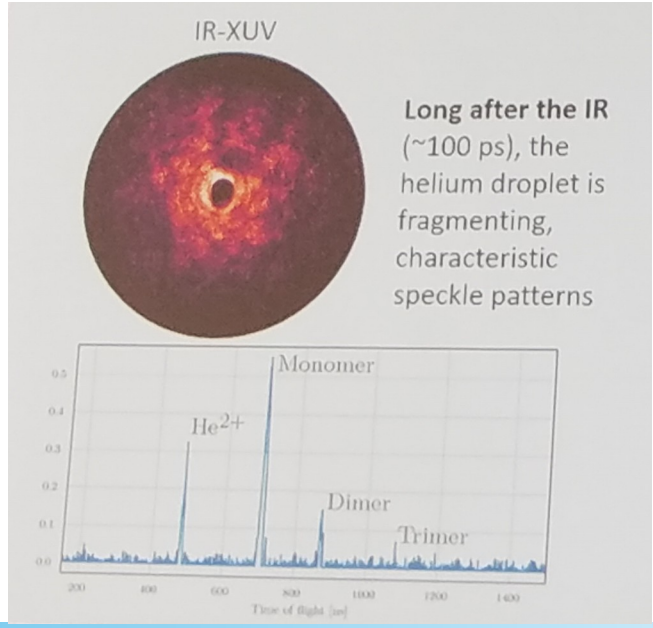
- Orientation by electric fields allowing separation of structural isomers and cluster sizes
- Focusing and separation of very large (bio)molecules
- Fixing molecular and laboratory frames for imaging:



Spectroscopy & imaging

Ultrafast dynamics in helium nanodroplets (Daniela Rupp):

- Time-resolved light scattering (Mie theory) for imaging of nano-scale objects (nanoparticles, helium nanodroplets)
- Determination of 3-D shapes of pure helium droplets, dynamics of helium droplet fragmentation



Spectroscopy & imaging

Polar molecules trapped in helium nanodroplets: electric field deflection, size separation, charge migration (Vitaly Kresin):

- Deflection of helium droplets with solvated polar molecule
- Measurement of dipole moment of the dopant
- Polar assemblies: monomers, dimers, trimers, etc. deflect differently
- Neutral droplet size separation by deflection

Spectroscopy & imaging

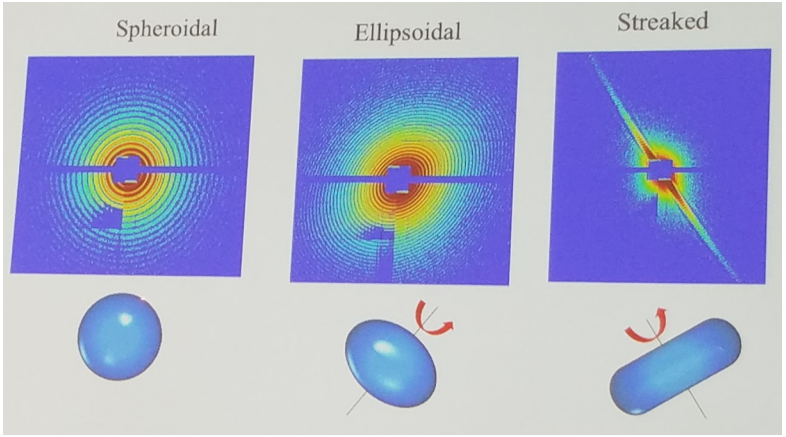
Intermolecular decay mechanism in doped helium droplets induced by XUV radiation (Aaron LaForge):

- Helium droplet enhanced double ionization of dopants
- Ionization of He atoms in the droplet result in double ionization of the dopant (Mg_n; ETMD)
- Resonant excitation of helium droplet produces double ionization of the dopant (alkali dimers; dICD)
- Model system for condensed matter ionization processes

Spectroscopy & imaging

Shapes of rotating ^{3}He droplets (Swetha Erukala):

- X-ray scattering image analysis of non-superfluid ^{3}He droplets (same method as used previously for ^{4}He droplets)
- Various shapes observed: Spheroidal, Ellipsoidal, Streaked:



- No trapping of Xe in vortices but they accumulate in ring on the outer wall of the droplet?

Spectroscopy & imaging

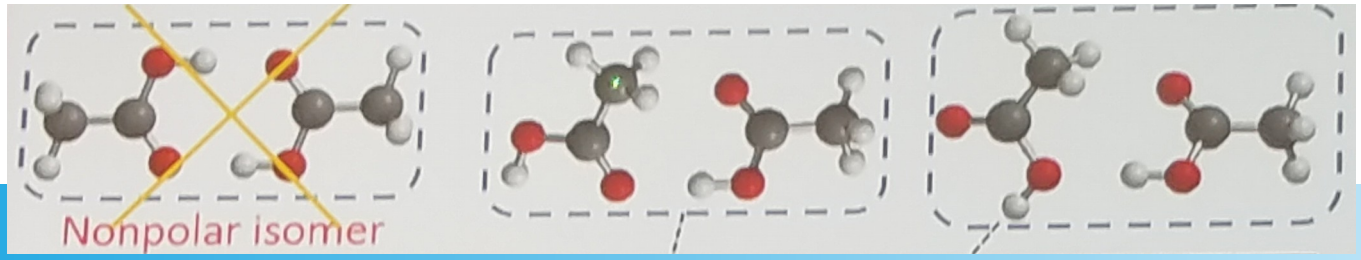
Accessing different binding sites of a multifunctional molecule in helium droplets (Devendra Mani *et al.*):

- Complex formation between propagyl alcohol and water within helium droplets
- Combined electronic structure and IR spectral analysis shows that the two structures formed predominantly are NOT the lowest energy configurations
- Long-range dipole-dipole interactions dominate the complexation

Spectroscopy & imaging

Accessing challenging molecular species using helium droplets: clusters, complexes, and ions (Andrew Ellis et al.):

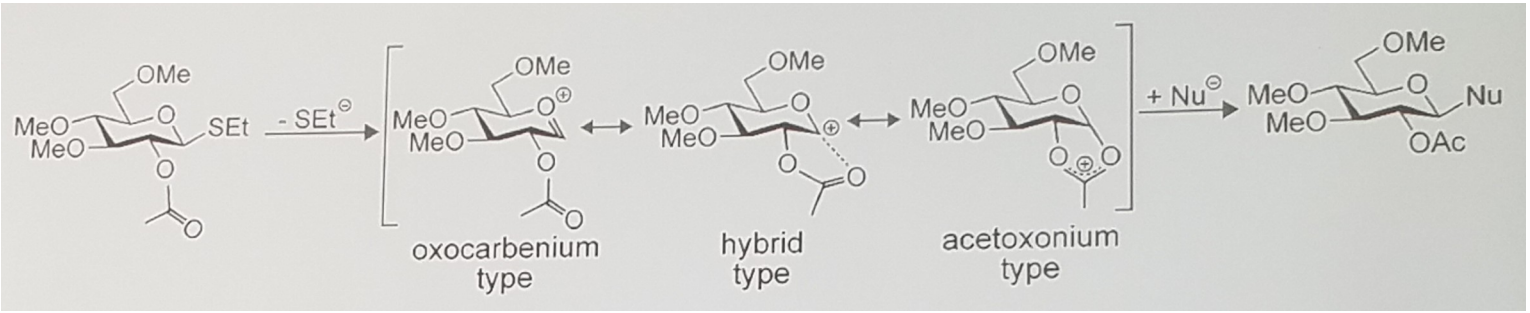
- IR spectroscopy of acetic acid and formic acid dimer and trimer cations in helium droplets
- Identification of their structures by combining electronic structure calculations
- Some surprises, e.g., the lowest energy structures not seen?



Spectroscopy & imaging

Spectroscopy of mass/charge selected cations and anions in helium droplets (Gert von Helden et al.):

- Fluorine chemistry - reactions with small molecules
- IR depletion spectroscopy of mass selected cations and anions
- Thermochemical access using thermalized ion trap
- Analysis of complex stereo-selective reactions:



Spectroscopy & imaging

Photoinduced formation of RbSr molecules on helium droplets from spatially separated Rb and Sr atoms (Florian Lackner et al.):

- R2PI spectroscopy of Sr, Sr_2, and RbSr on helium nanodroplets
- Photo-induced formation of SrRb and Sr_2 on the droplet (?)
- Evidence for Sr being inside AND outside the droplet
- How common phenomenon is this? Only very few examples so far?

Spectroscopy & imaging

Infrared spectroscopy of alkyl radicals in helium droplets and solid para-hydrogen (Gary Douberly et al.):

- Characterize radicals relevant to low-temperature combustion
- Mass and IR spectrometry of pyrolytic decomposition of organics (n-propyl and i-propyl)
- Low temperature of helium droplets allows for high-resolution IR
- Local model Hamiltonian approach with possible empirical prescription for some of the off-diagonal elements

Spectroscopy & imaging

Acid solvation versus dissociation at "stardust conditions":

reaction sequence matters! (Martina Havenith et al.):

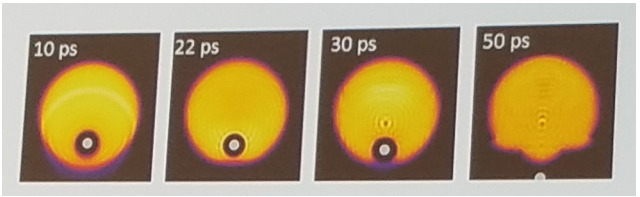
- Hydrogen bonding of water using IR spectroscopy
- Solvation of HCl in water clusters vs. deprotonation of HCl?
- Deprotonation at four water molecules but this depends on the order: adding acid to water or adding water to acid (deprot)

"Adding water to acid results in a splash!"

Spectroscopy & imaging

Femtosecond photoexcitation dynamics of atoms and molecules inside helium nanodroplets (Markus Koch et al.):

- Dynamics of In and In_2 in helium droplets
- In resides inside the droplet and excitation opens up a bubble (30 ps) but is then ejected from the droplet:
- Wavepacket dynamics of In_2 inside the droplet (dissipative environment)



Spectroscopy & imaging

Stark-spectroscopy investigations of large organic molecules in superfluid helium nanodroplets (Alkwin Slenczka et al.):

- Stark spectroscopy of large organic molecules
- Aim at the zero phonon line and identify the rotational structure
- Demonstration of the method by observing the ZPL intensities
- Mystery: Adding one H_2O molecule changes the Stark response of the ZPL?

Spectroscopy & imaging

Auger emission from the Coulomb explosion of helium nanoplasmas (Josef Tiggesbäumker et al.):

- Coulomb explosion of helium droplets: fast electrons, energetic ions in high charge states, xrays
- Charging Xe clusters inside droplets and pure droplets
- Observation of Auger emission from plasma with structure corresponding to one-electron He^+

Spectroscopy & imaging

Quantum vortices in superfluid helium droplets (Andrey Vilesov):

- Introduction to quantum vortices (also stressing curl $v = 0$)
- Connection between vorticity and droplet shapes (capillary waves important in prolate droplets)
- Trapping of impurities by vortices and helium droplets
- Detection of vorticity in droplets by ultrafast xray and XUV tech.
- Analysis of scattering images

Spectroscopy & imaging

Serial single molecule electron diffraction imaging: a journey

(Wei Kong):

- Serial single molecule electron diffraction imaging
- Methods for determining molecular structure (proteins)
- New method for determining structure without single crystals
- Diffraction pattern of (rotating) ferrocene, molecular iodine + dimer (structure), pyrene in droplets (monomer, dimers)
- Proteins in droplets and alignment (aniline; suppressed ionization; coexistence of two cations in one droplet)

Spectroscopy & imaging

Ultrafast energy- and charge-transfer in He nanodroplets studied by femtosecond XUV and X-ray techniques (Oliver Gessner):

- Xray and XUV excitation of helium droplets (pure and Rg doped)
- Complex electron-nuclear dynamics after excitation (pump-probe)
- Mechanism of solute-solvent energy and charge transfer processes
- Unentangle the dynamics by XUV + UV femtosecond spectroscopy
- Time-resolved imaging of strong-field induced cluster dynamics

Spectroscopy & imaging

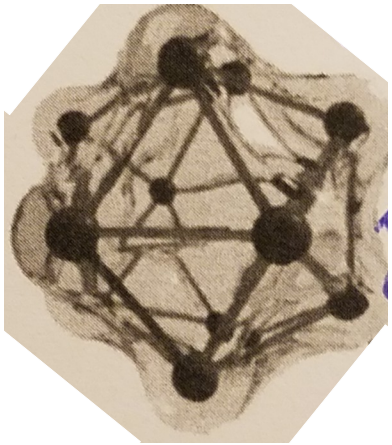
Strong-field nanophysics: new routes to imaging the classical and quantum dynamics of finite systems (Thomas Fennel):

- Classical and quantum aspects of light-matter interactions by multicolor pump-probe spectroscopy (clusters in strong fields)
- Characterization of XUV induced plasma by “nanoplasma oscilloscope” - XUV multistep ionization (Ar clusters; MD simul.)
- Quantum coherent diffractive imaging (CDI): spatiotemporal dynamics by XUV (1s-2p resonant)

Novel structures & solvation

Multiply charged helium droplets (Paul Scheier *et al.*):

- Controlled production of large droplets with $-6 \leq Z \leq +30$
- Charged centers in the droplet provide growth centers for dopants with size tuning (and possibly route for new chemistry!)
- Application also to Helium tagging spectroscopy

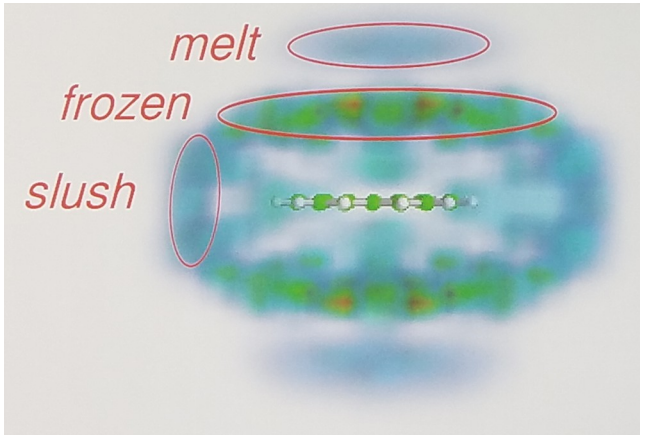


"Delicate balance between repulsion between the charges and attraction to the droplet"

Novel structures & solvation

Stepwise solvation of polycyclic aromatic hydrocarbons by helium: curvature and dynamical effects (F. Calvo *et al.*):

- First solvent shell is strongly localized (near hexagonal & pentagonal sites)
- The following shells are less localized
- Stabilization of helium between dimers? (solvent shell effects; “quantum gels”)
- However, the effect of exchange?



Novel structures & solvation

Experimental characterization of low-temperature surface reations (Serge Krasnokutski *et al.*):

- Determination of lowest energy structures based on evaporation of helium from droplets (“nanocalorimeter”; pressure monitoring)
- Data can be compared directly with energetics from *ab initio* calculations (identification of products)
- First observation of C_2O_2 – structure was non-linear!

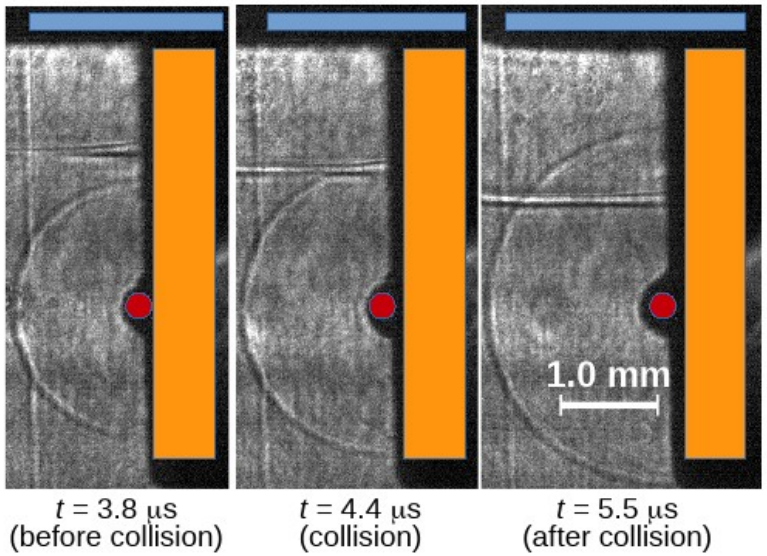
Novel structures & solvation

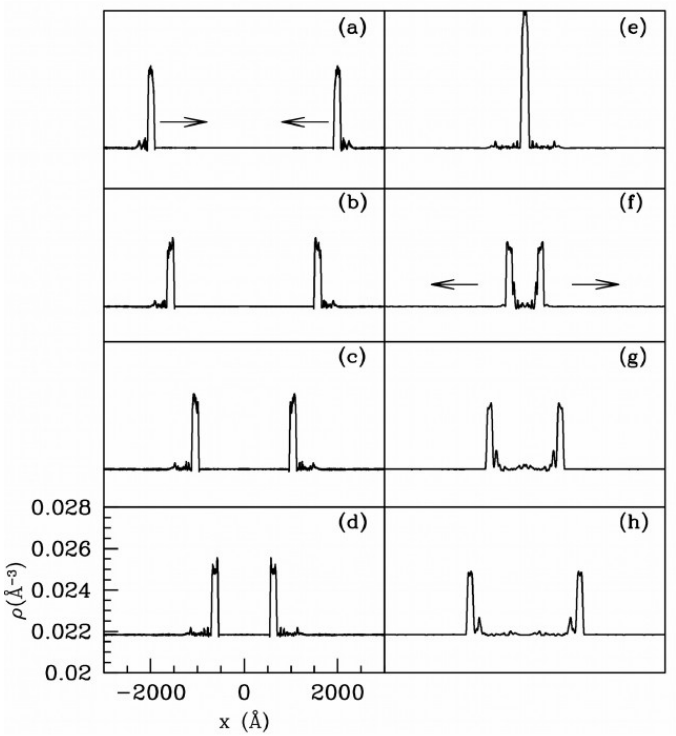
Influence of electron scattering on the properties of the hydrated electron (Ruth Signorell):

- Electron scattering from water clusters by angle-selective photoemission
- Determine the differential scattering cross-section as a function of energy and determine the electron binding energy
- Goal: solvation structure of the electron in water and the existence of long-lived surface states

What have I been up to recently?

First observation of bright solitons in superfluid ^{4}He:



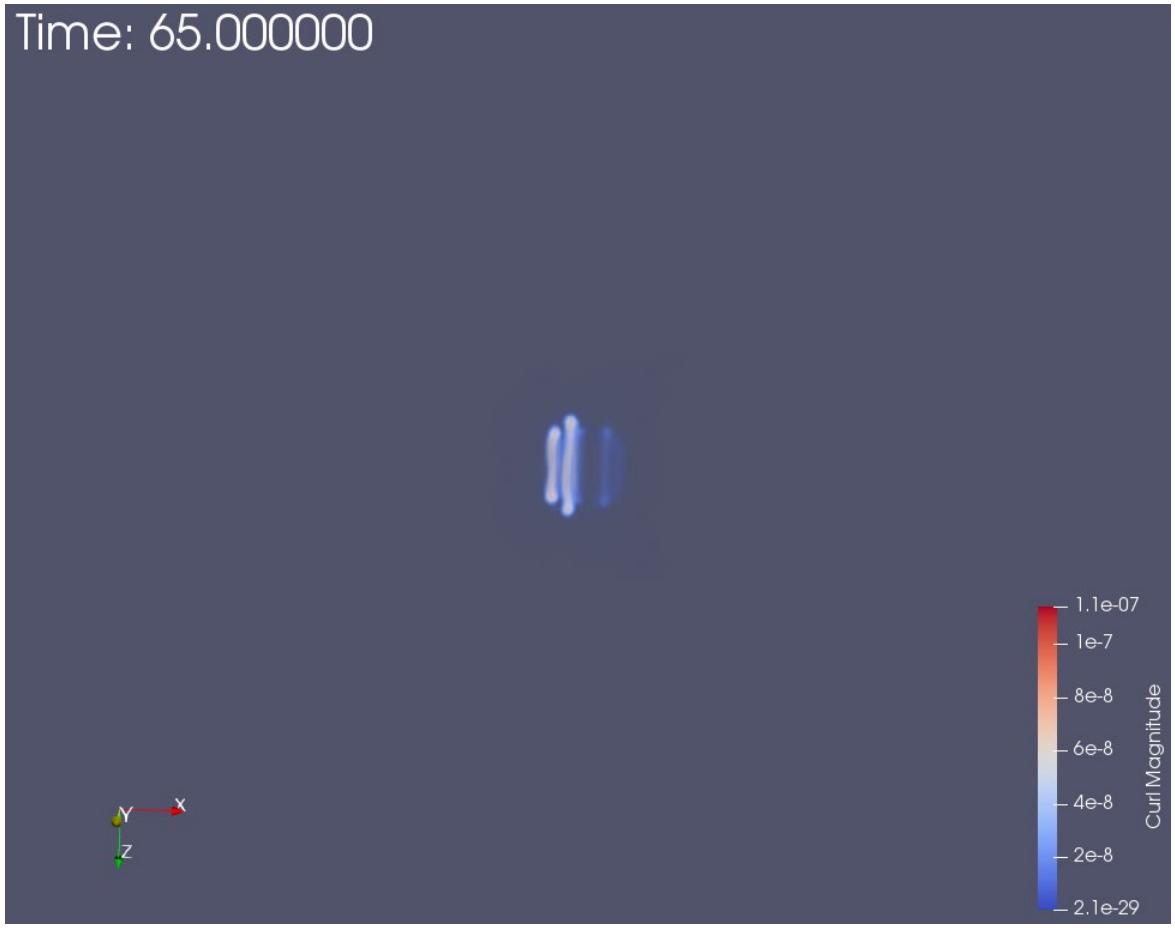


TDDFT simulation

Observation: Phys. Rev. Lett. 120, 035302 (2018)
Soliton trains: Phys. Rev. B 99, 144508 (2019)

And on quantum vorticity/turbulence:

GPU implementation
of He-TDDFT



Flow past a sphere above the Feynman critical velocity

Phys. Rev. B 98, 094520 (2018).

BIG THANKS TO THE ORGANIZERS!!!!!!!!