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Femtosecond Photoexcitation Dynamics of Atoms and Molecules inside Helium Nanodroplets

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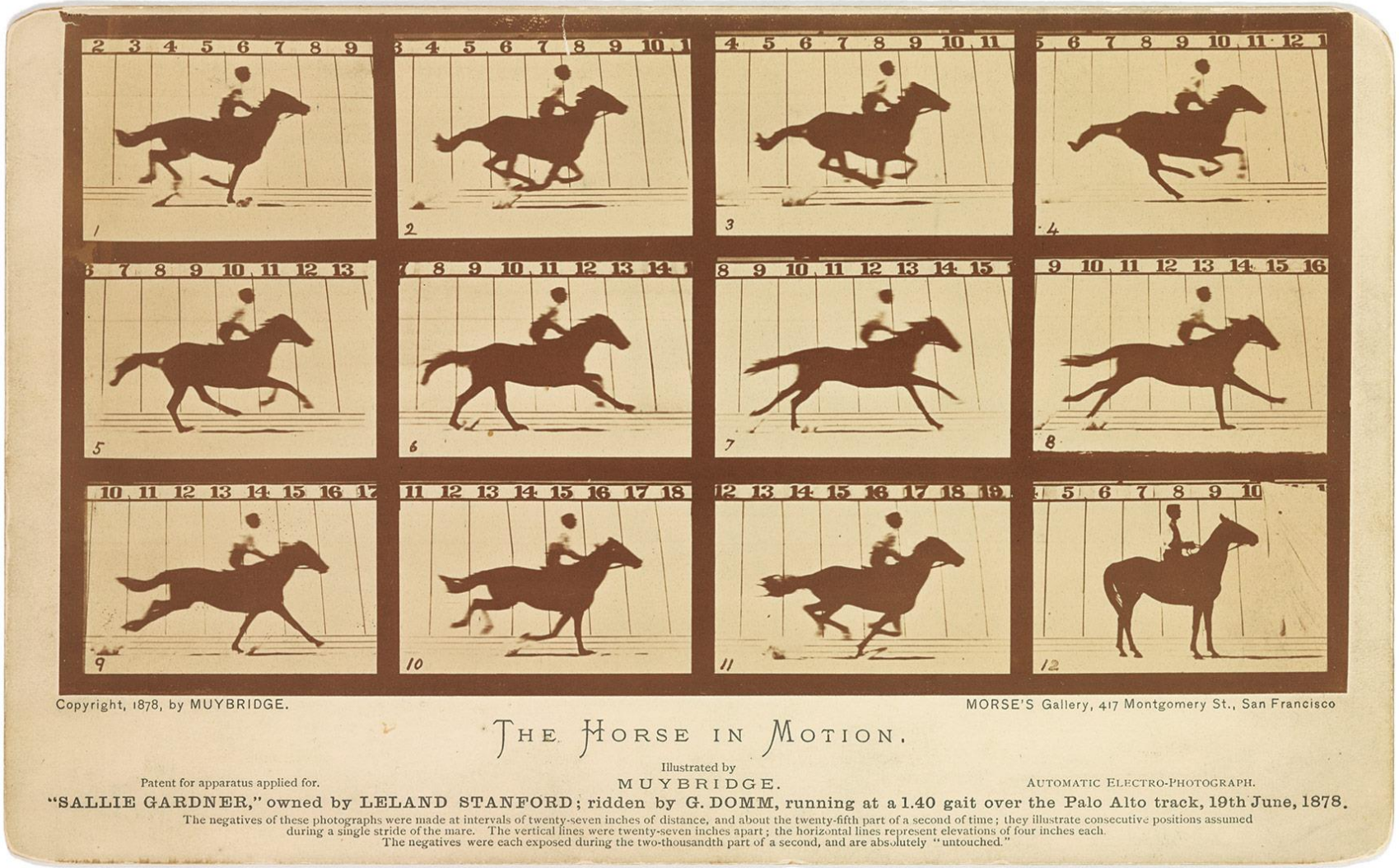
Graz University of Technology,
Institute of Experimental Physics

Quantum Fluid Clusters 2019
Bad Honnef, May 21st, 2019

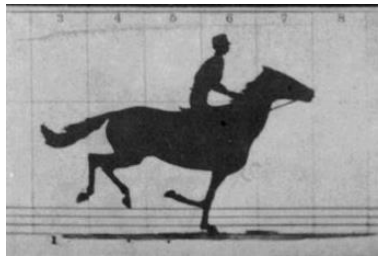
The Horse in Motion

Eadweard Muybridge 1878



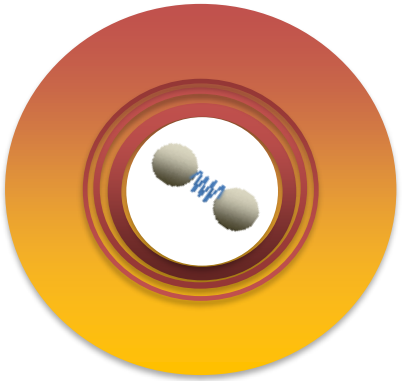
The Molecule in Motion

(inside a He droplet)



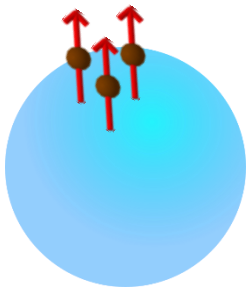
1878

2019



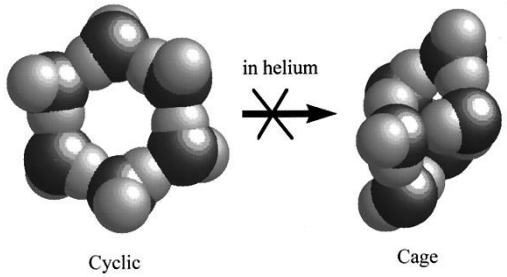
Helium nanodroplet experiments

Fragile species: $^4Na_3$



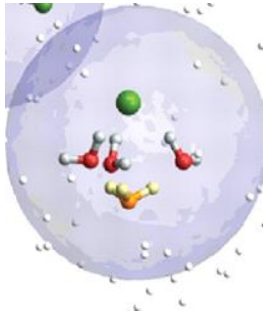
*Science **273***, 629 (1996)

The smallest piece of ice: $(H_2O)_6$



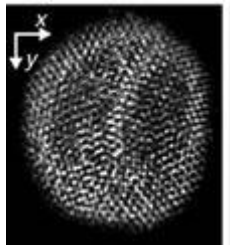
*Science **287***, 293-295 (2000)

Microsolvation: The smallest drop. of acid: $HCl(H_2O)_4$



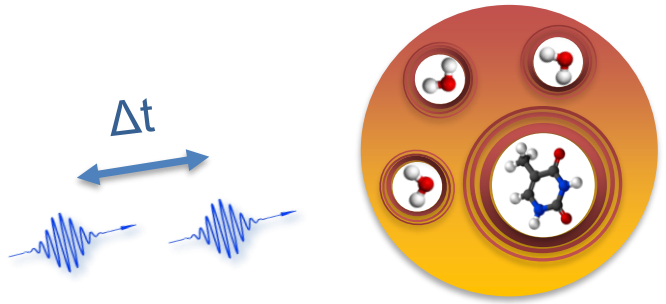
*Science **324***, 1545-1548 (2009)

Core-shell clusters



*Nat Commun **6***, 8779 (2015)

Femtochemistry inside He_N

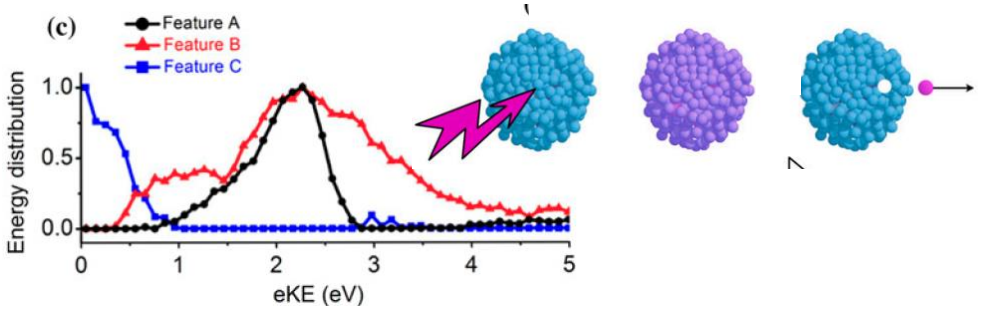


Ahmed Zewail

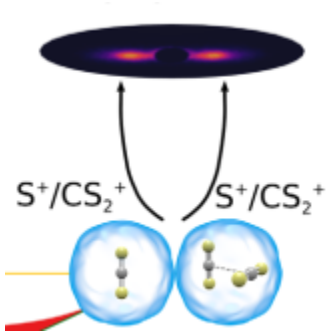
Time-resolved He_N experiments

Pure droplets: Daniel Neumark, Oliver Gessner, et al.

Int. Rev. Phys. Chem. **34**, 239 (2015)



Alignment & rotation: Henrik Stapelfeldt, et al.



PRL 120, 113202 (2018)

Nat Commun 10, 133 (2019)

rotational revivals

PRL 118, 203203 (2017)

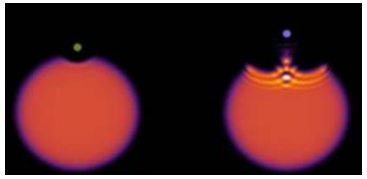
Surface-located alkali-metals:

Frank Stienkemeier, Marcel Mudrich , et al. (Int. Rev. Phys. Chem. **33**, 301, 2014)

Simulations: Manuel Barranco, Marti Pi , et al.

atoms:

observe desorption in real time (1-100 ps)



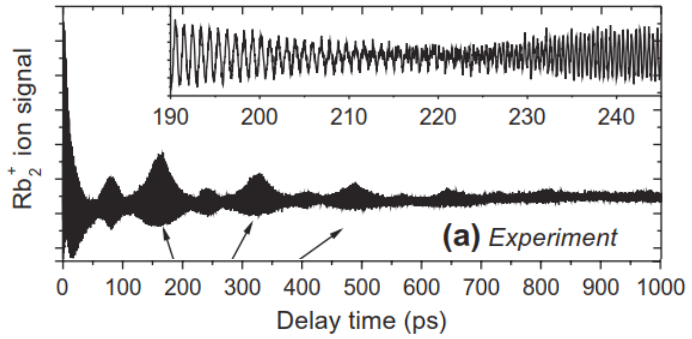
JPCL 8, 307

molecules: coherent vibrations

Rb_2: 1.5 ns

PRA 80, 042512 (2009)

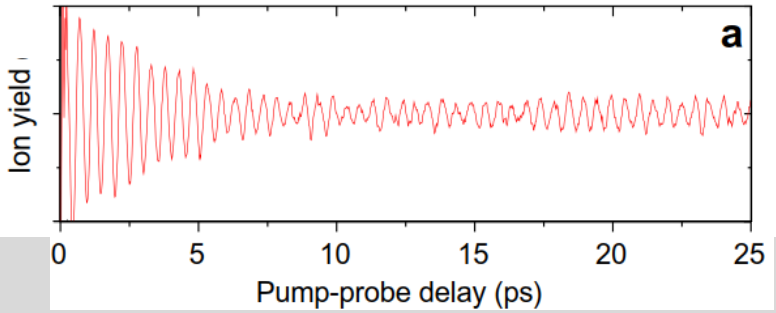
PCCP 13, 6816 (2011)



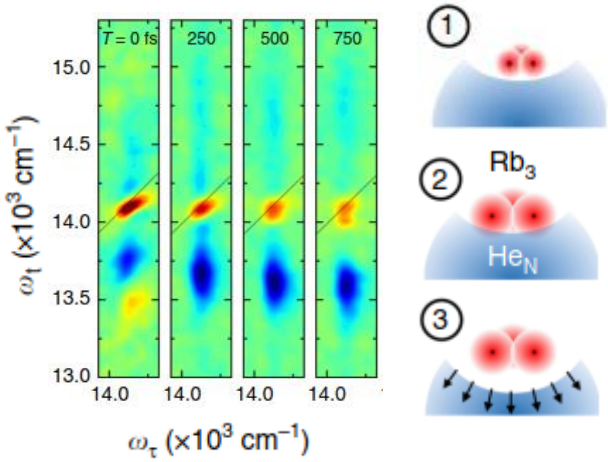
K_2: ~5 ps

J Phys B 39, S1151 (2006)

CPL 490, 245 (2010)



2D spectroscopy

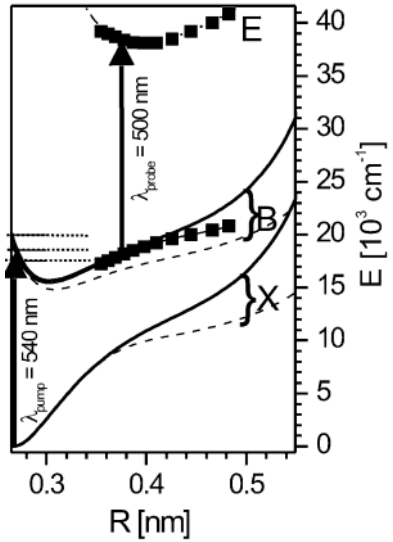


Nat Commun 9, 4823 (2018)

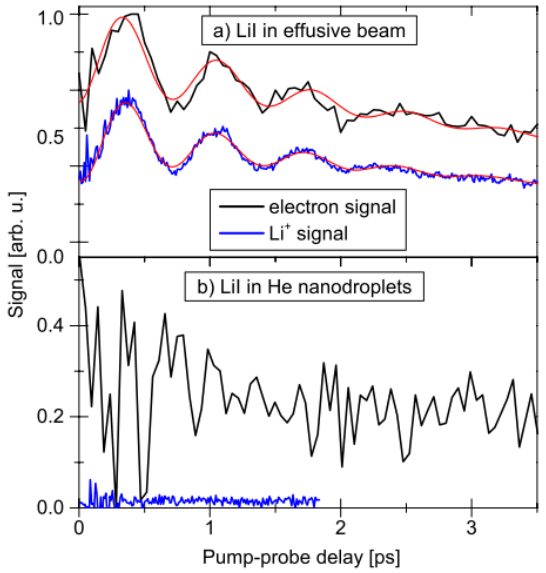
Femtochemistry inside He_N: open questions

Influence of droplets on intra-molecular dynamics?

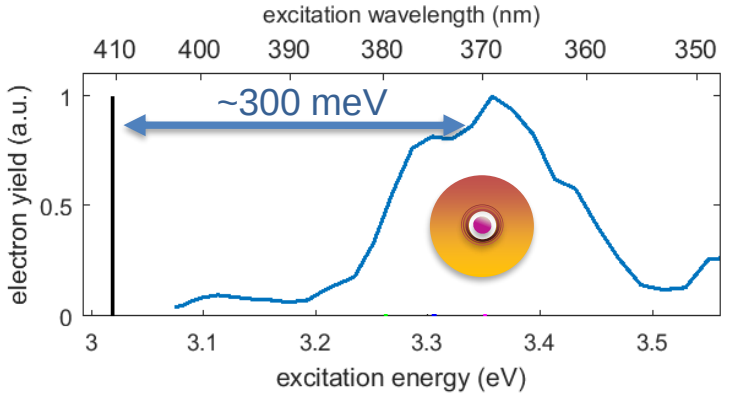
I_2:Kr PCCP 9, 779 (2007)



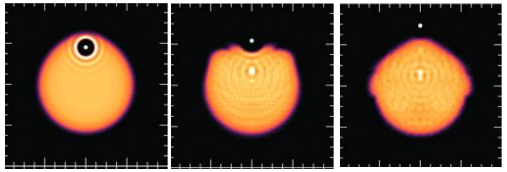
LiI-He_N JCP 142, 044303 (2015)



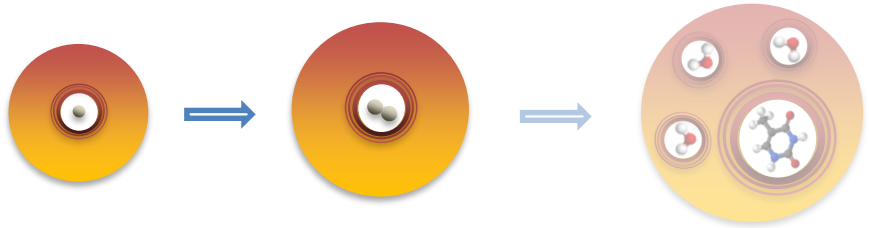
Droplet response to photoexcitation?



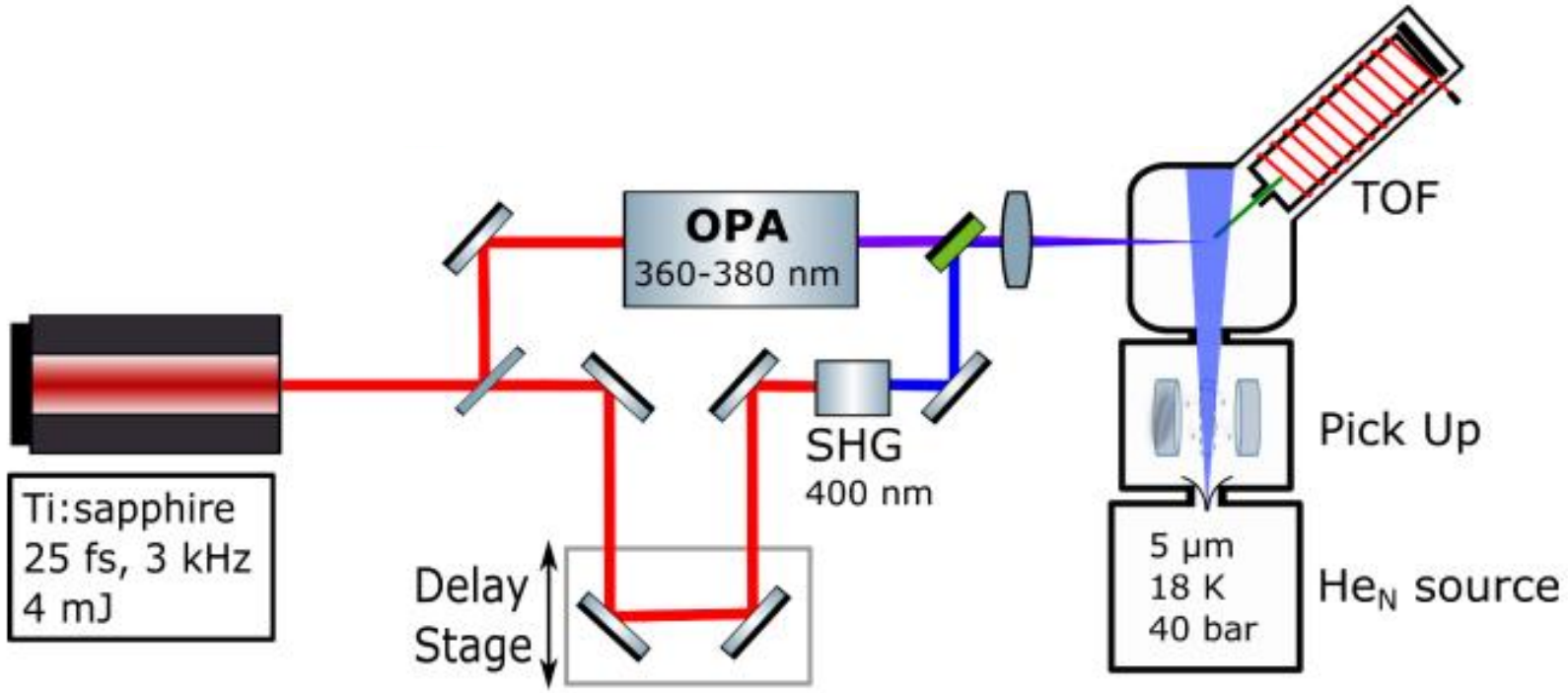
Accessible time frame?



Ag-He_N: PCCP 15, 18388 (2013)

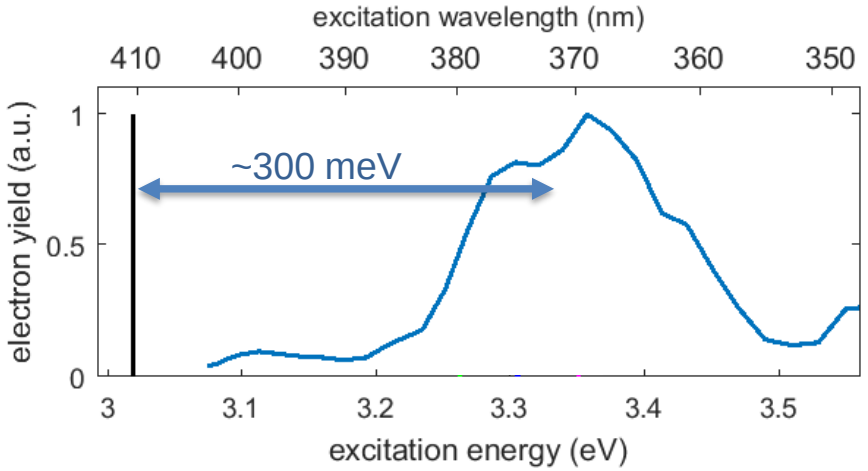


The experiment

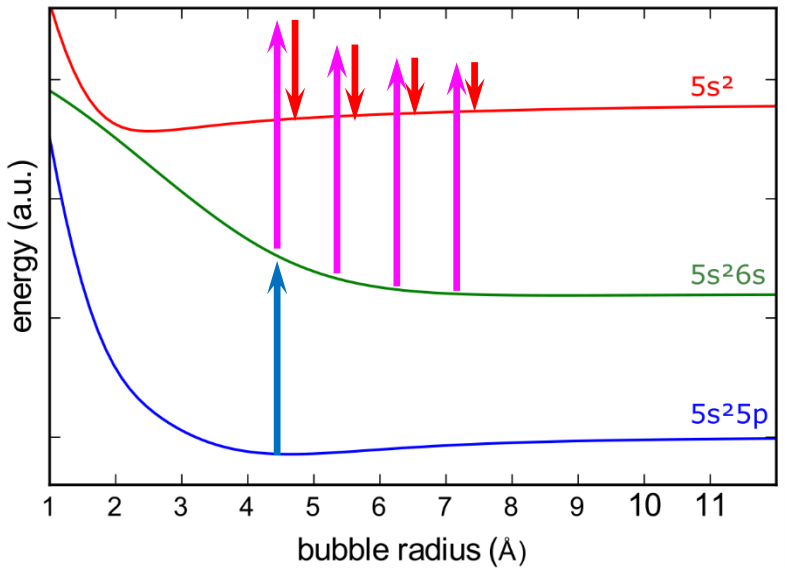


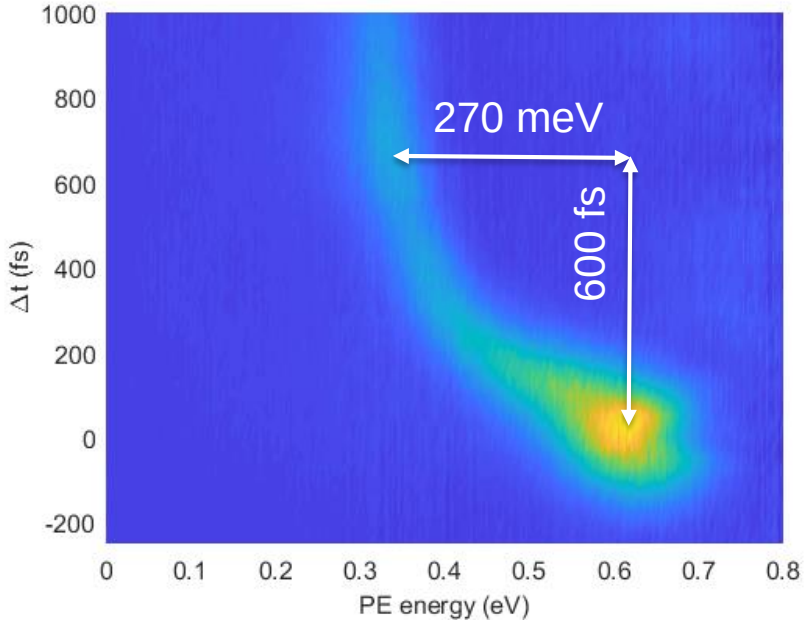
Droplet response to photoexcitation of atoms

In-He_N



B. Thaler et al.,
JPC A 123, 3977 (2019)





In-He_N: bubble expansion

Manuel Barranco

Marti Pi

Nadine Halberstadt

Time Dependent Helium-Density-Functional-Theory

BCN-TLS-HeDFT computing package: https://github.com/bcntls2016

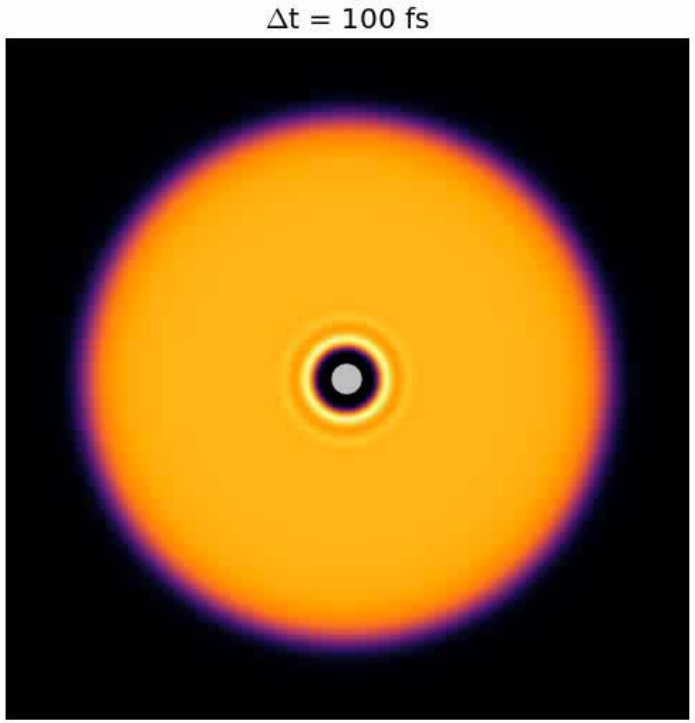
Andreas Hauser

Ralf Meyer

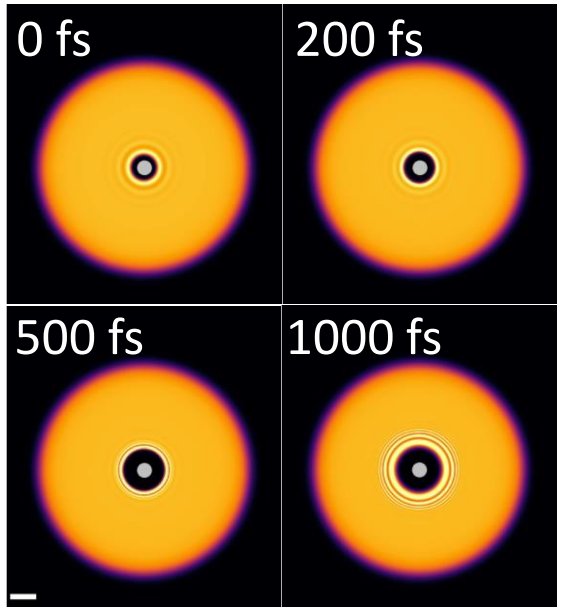
He density ρ_{He} as the functional parameter

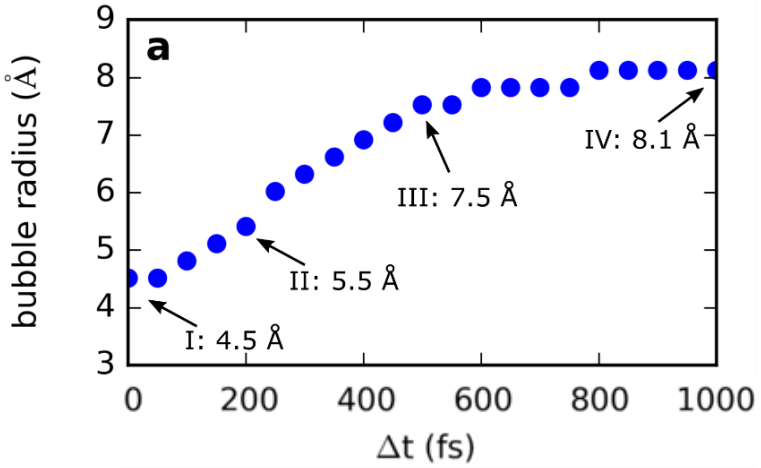
$\Rightarrow$ solve Schrödinger equation for ρ_{He}

$\Rightarrow$ solve equation of motion for the impurity

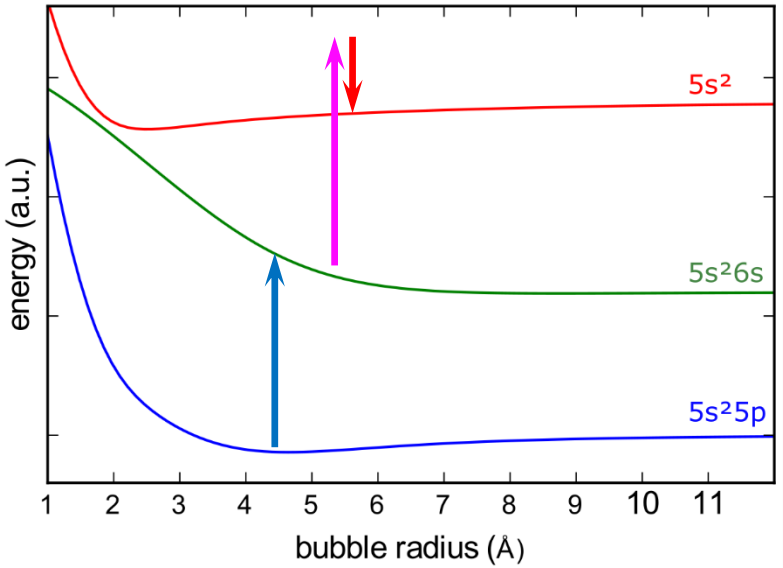


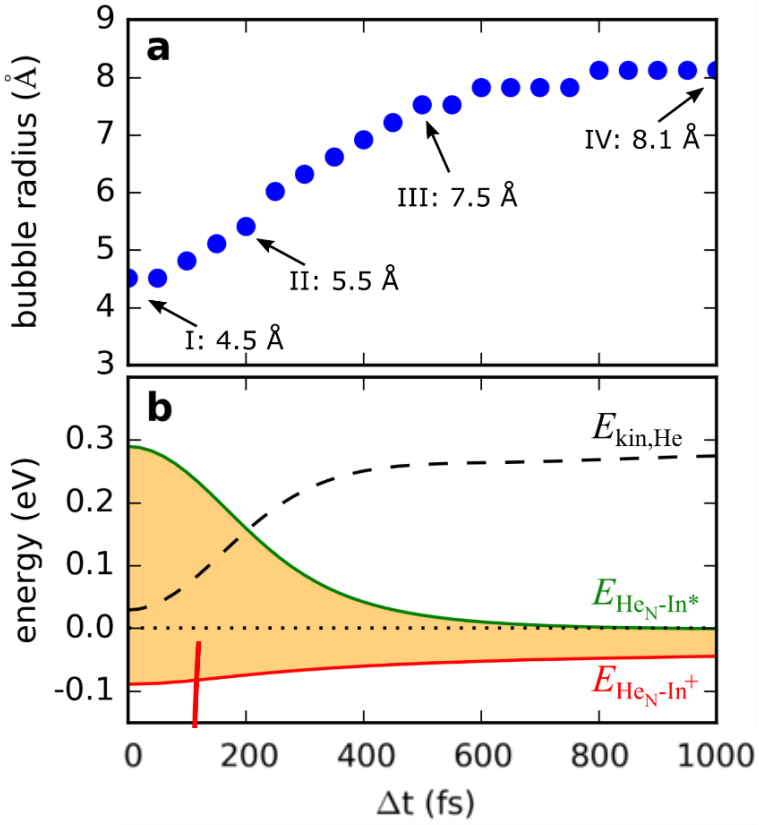
Energetics of bubble expansion



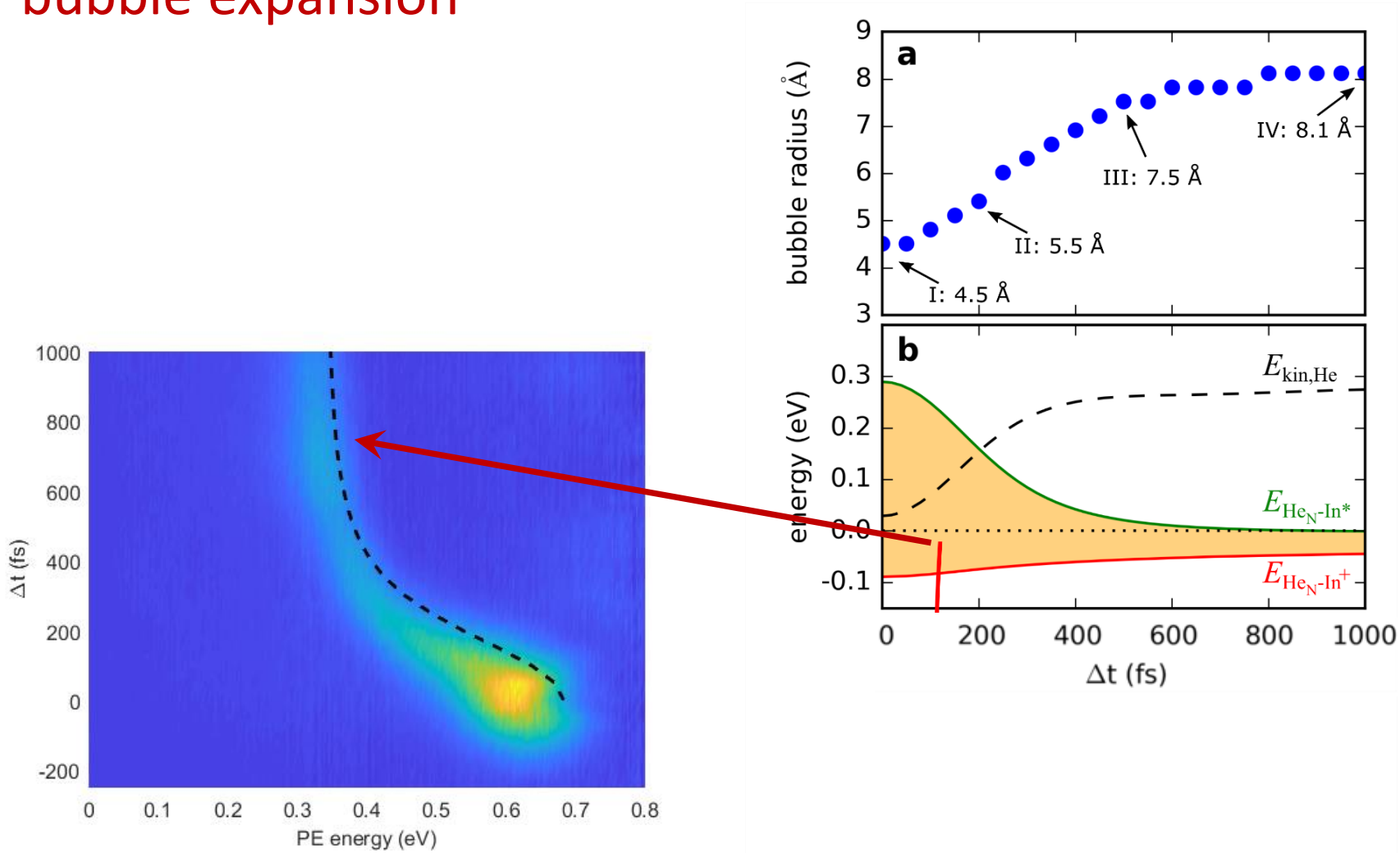


Energetics of bubble expansion

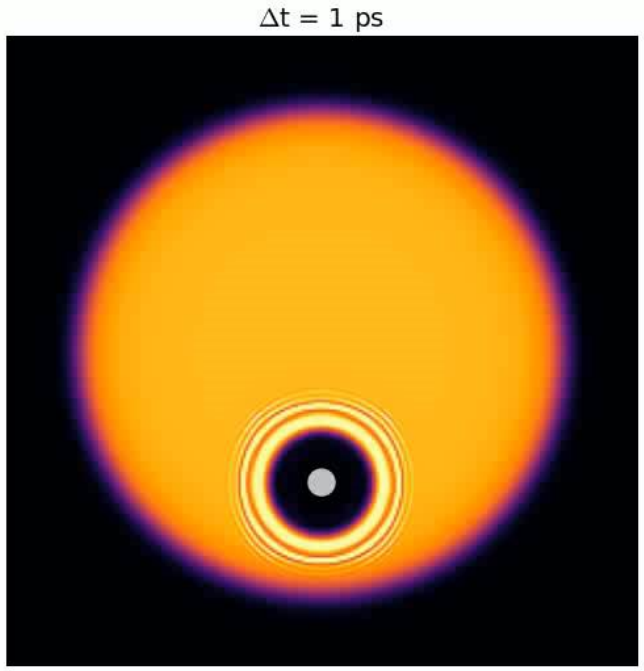


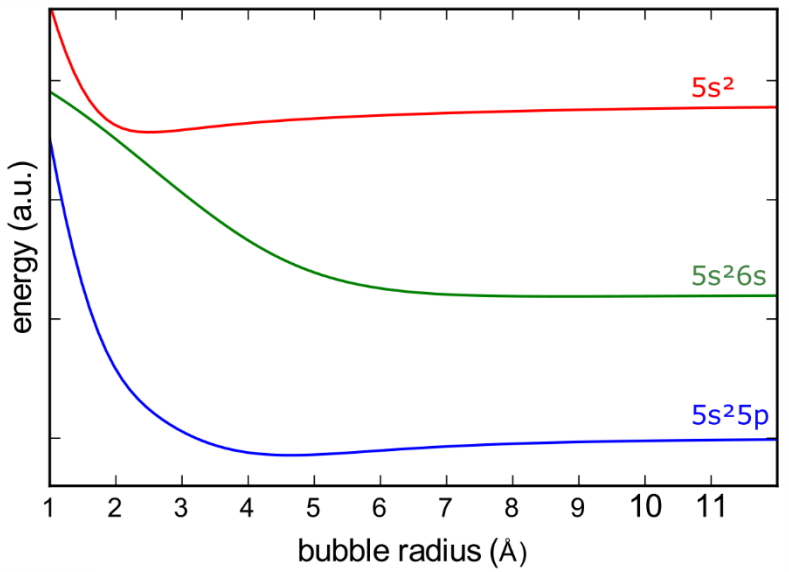


Energetics of bubble expansion

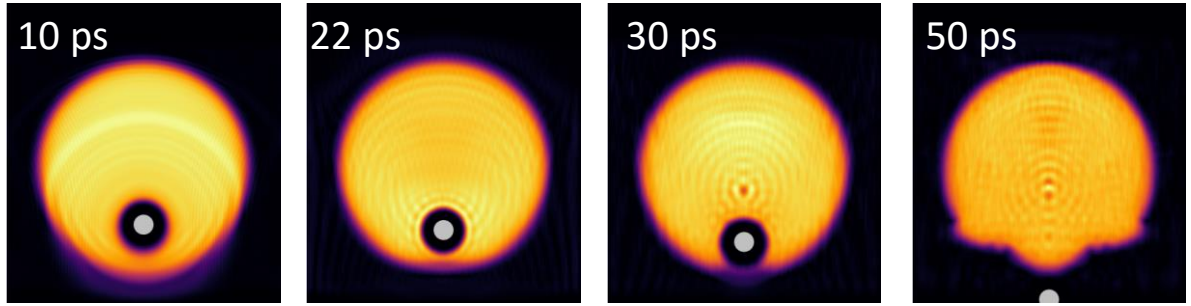


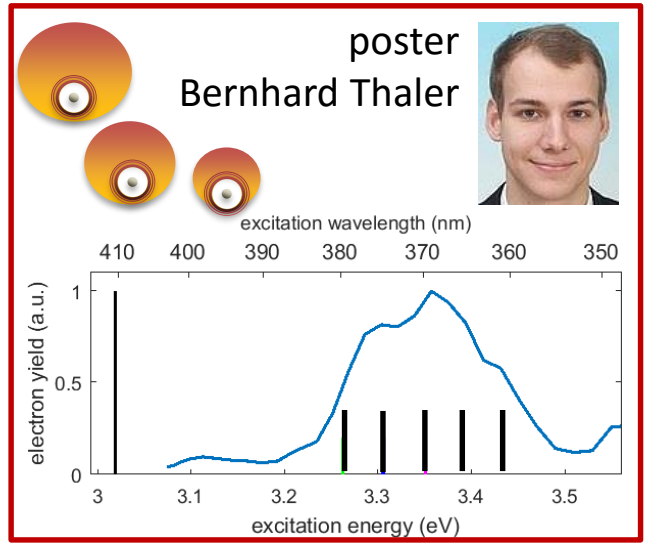
In-He_N: ejection and bubble oscillation

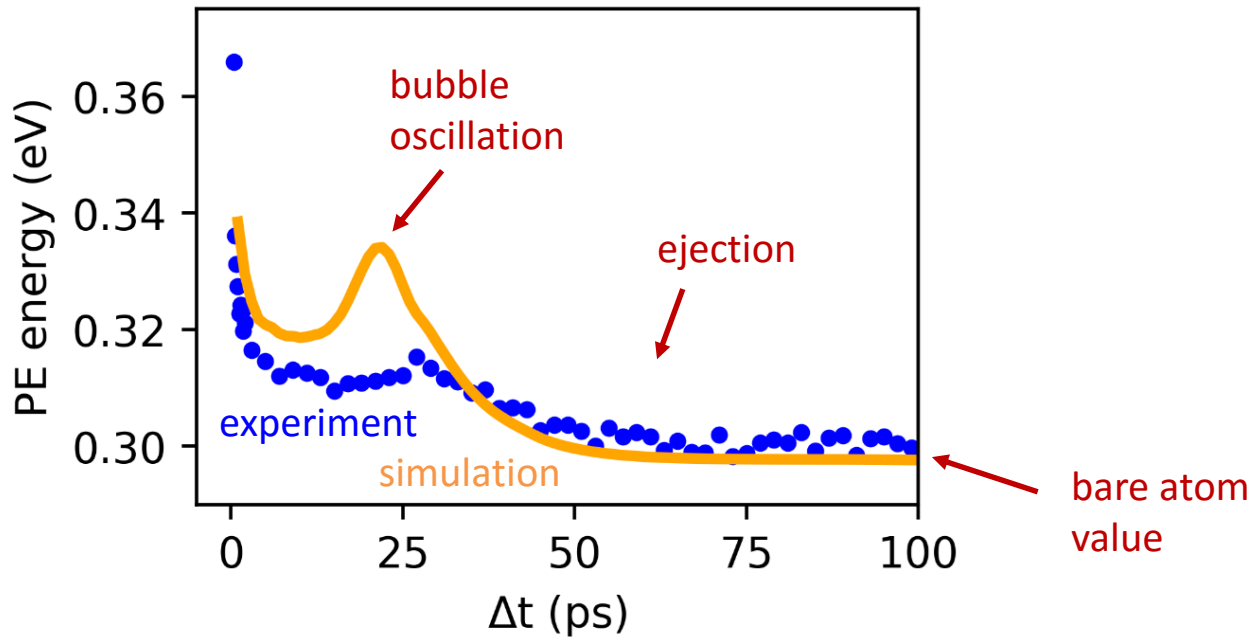


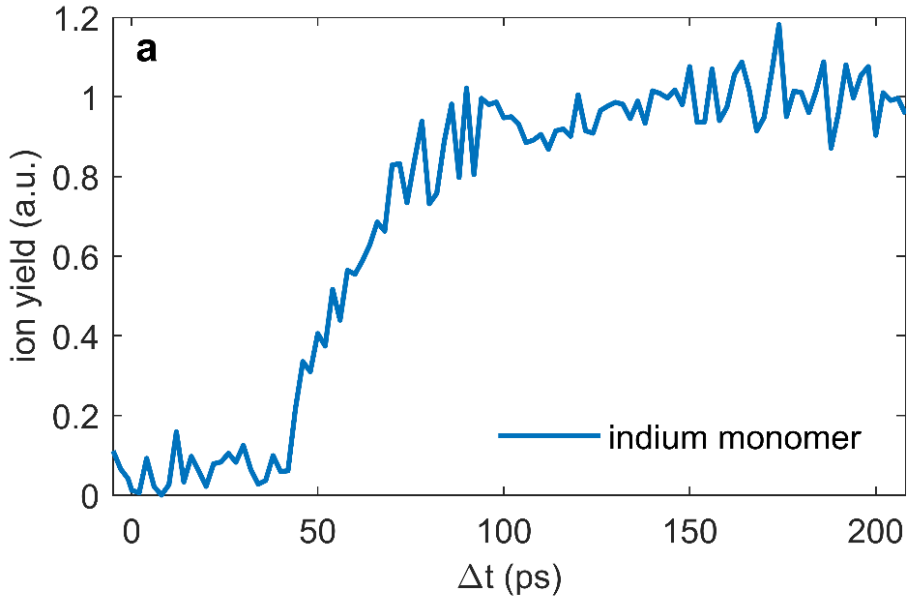


In-He_N: ejection and bubble oscillation



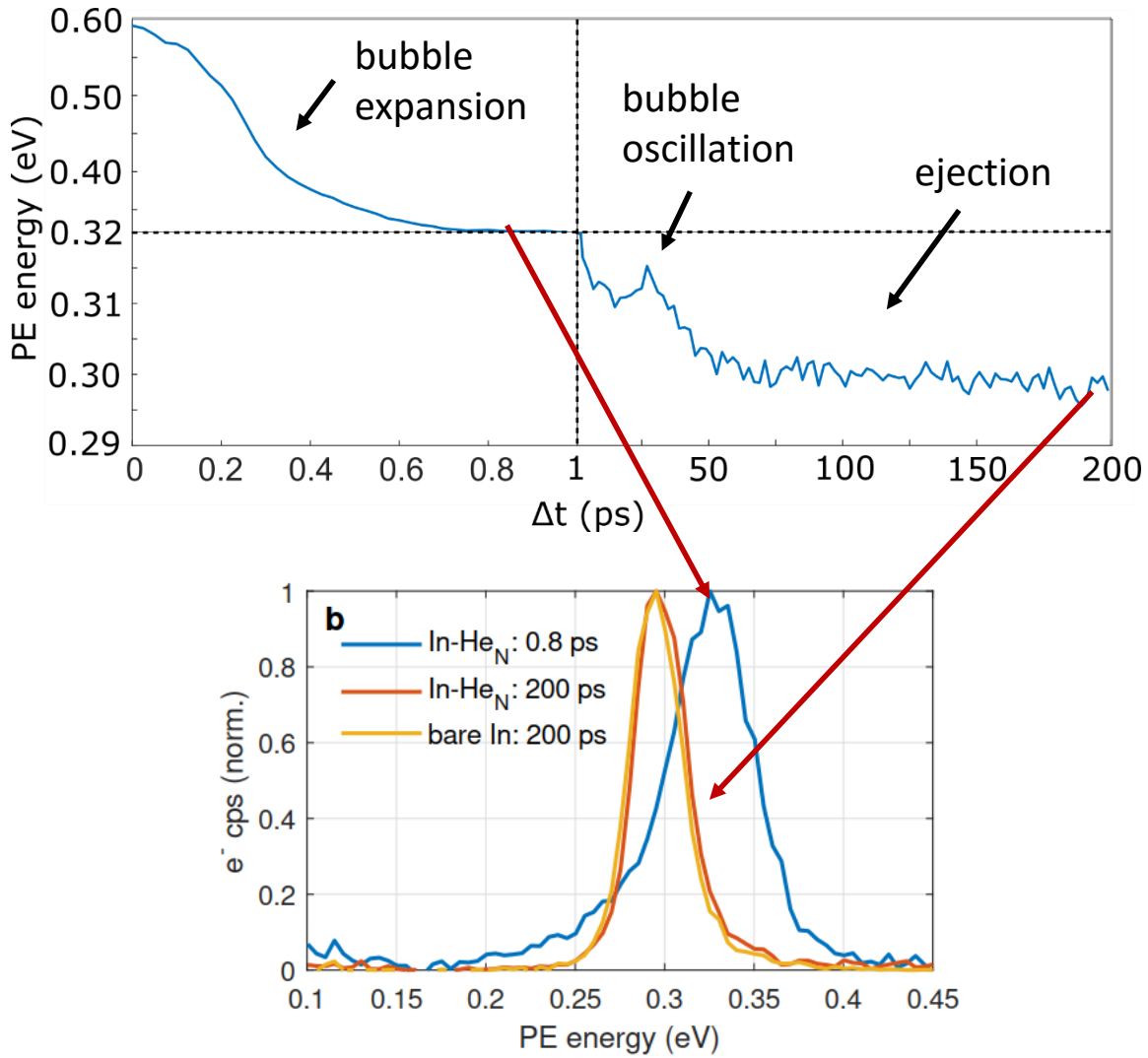




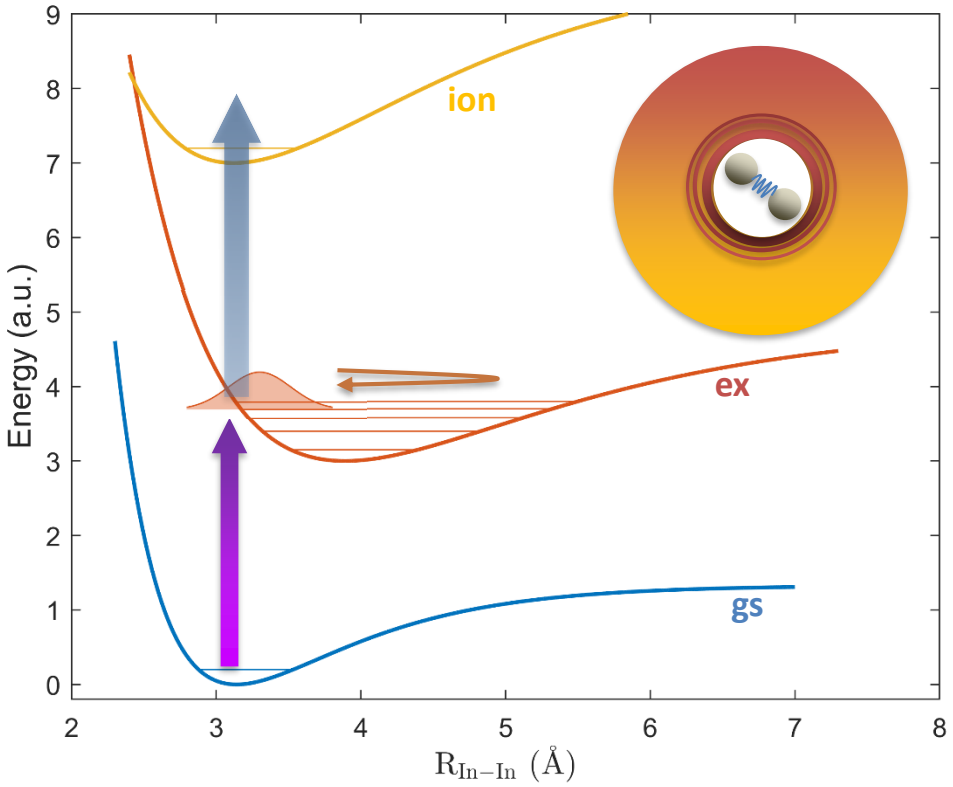


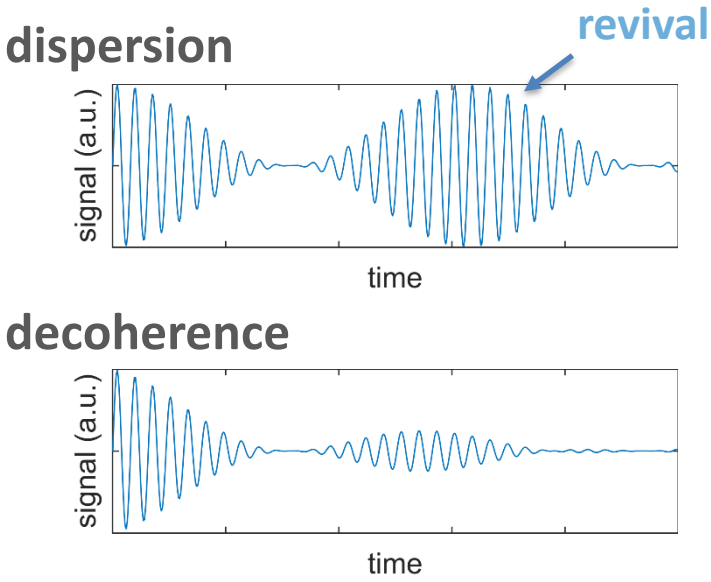
B. Thaler et al., Nat. Commun. **9**, 4006, 2018

Photoelectrons: sensitive observable

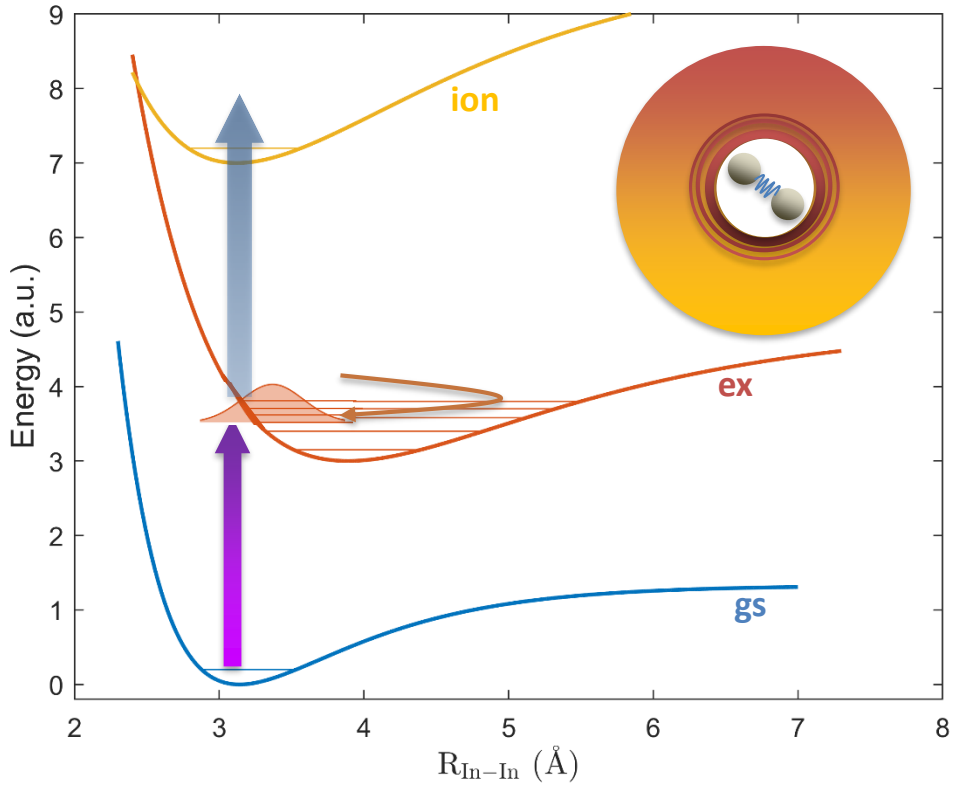


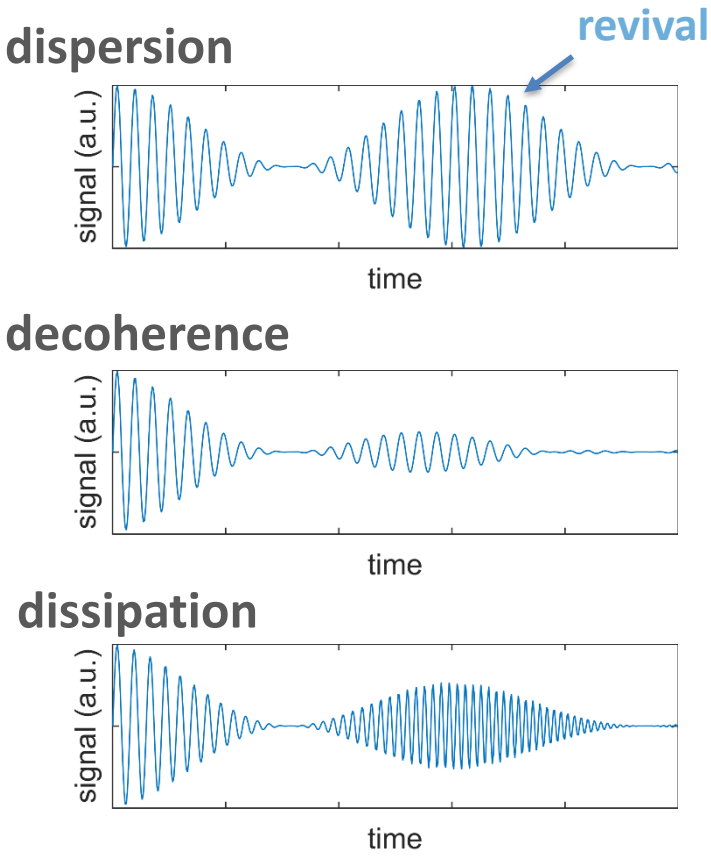
Wave packet dynamics in a dissipative environment





Wave packet dynamics in a dissipative environment

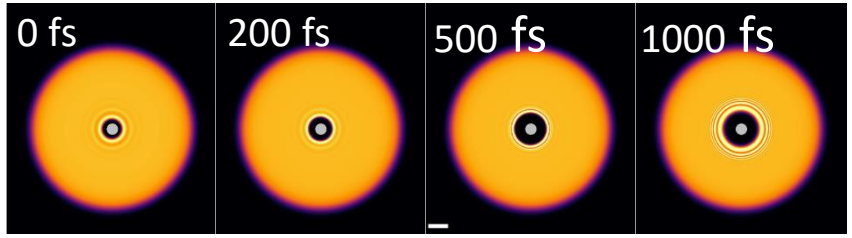




Summary – towards Femtochemistry inside He droplets

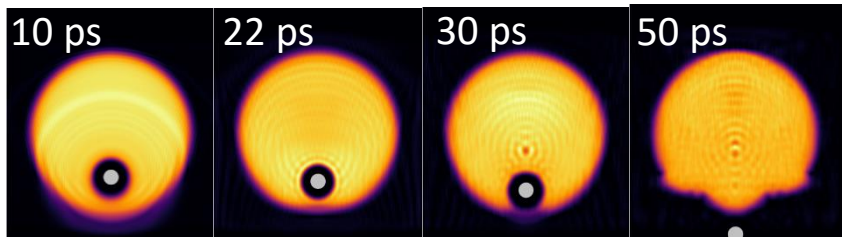
- ✓ photoelectrons are a good observable
- ✓ quantum solvent response to dopant photo excitation:

bubble expansion: ~600 fs



bubble oscillation: ~30 ps

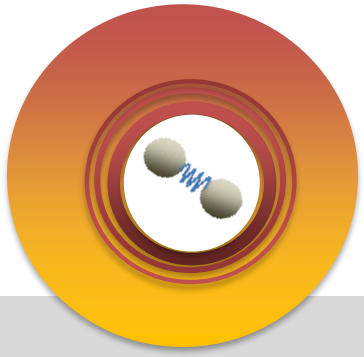
atom ejection: ~50 ps



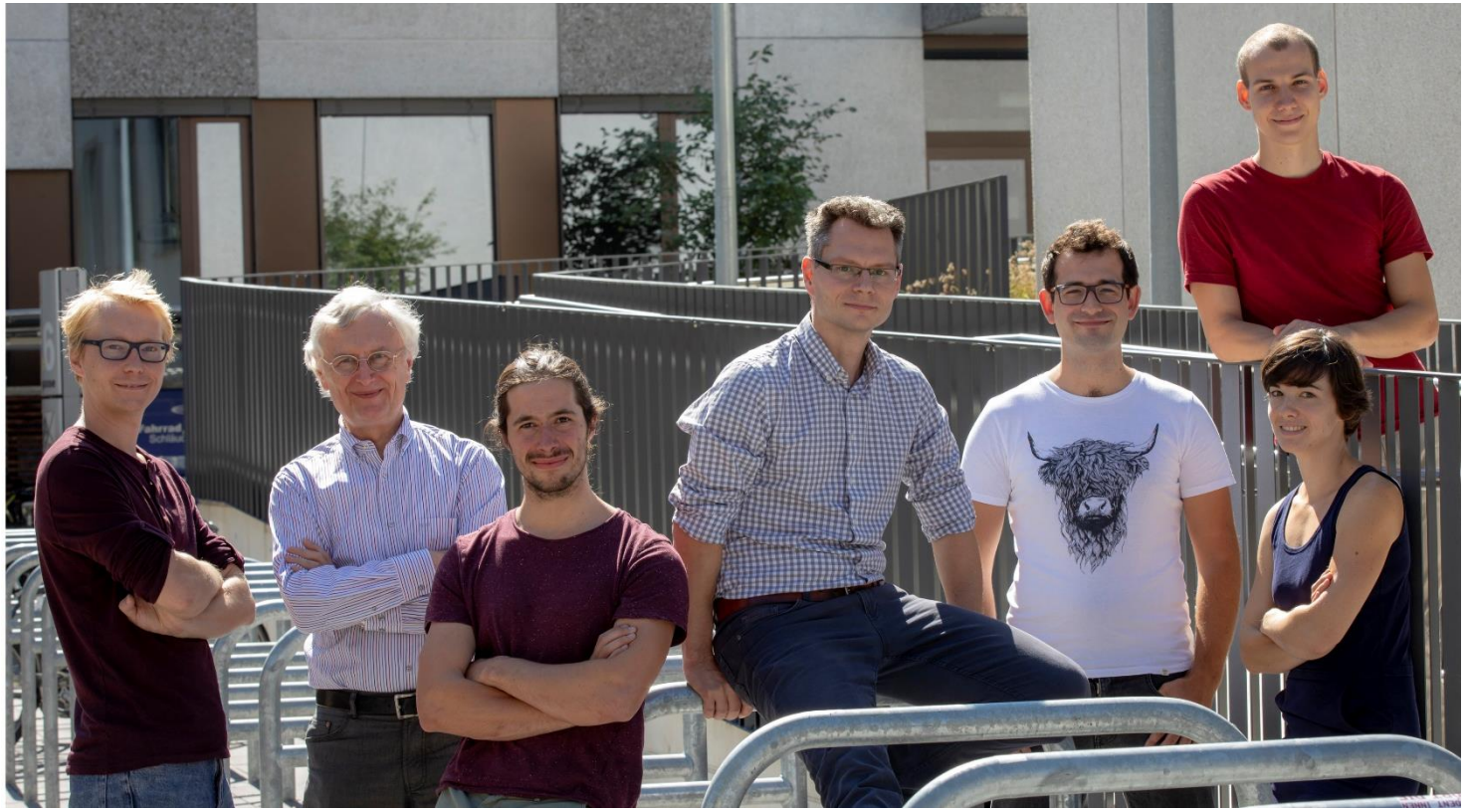
B. Thaler et al., Nat. Comm. **9**, 4006, 2018

- ✓ little influence on intrinsic dynamics (In_2)

He is an exceptional solvent:
long vibr. coherence (~100 ps)
(water: <1 ps)



Acknowledgements



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