Title: Observation of a molecular bond between ions and Rydberg atoms using a high-resolution pulsed ion microscope

Speakers: Tilman Pfau

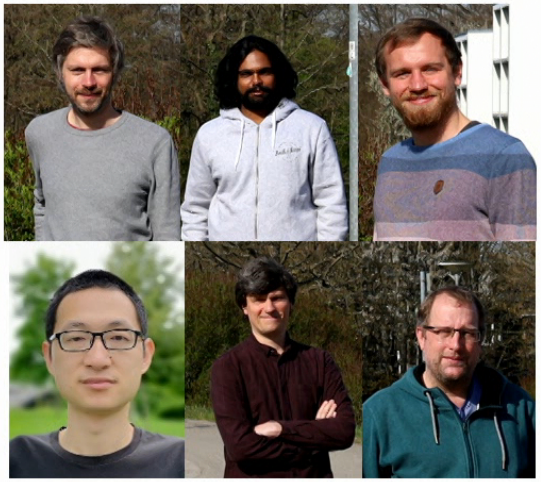
Collection: Cold Atom Molecule Interactions (CATMIN)

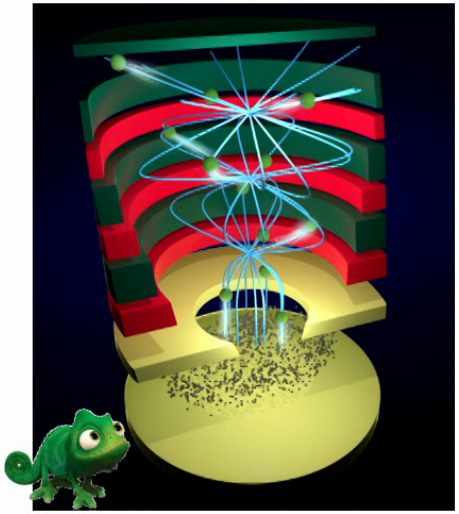
Date: July 14, 2022 - 9:00 AM

URL: https://pirsa.org/22070004

Abstract: "We present our recent studies on Rydberg atom-Ion interactions and the spatial imaging of a novel type of molecular ion using a high-resolution ion microscope. The ion microscope provides an exceptional spatial and temporal resolution on a single atom level, where a highly tuneable magnification ranging from 200 to over 1500, a resolution better than 200nm and a depth of field of more than 70µm were demonstrated [1]. A pulsed operation mode of the microscope combined with the excellent electric field compensation enables the study of highly excited Rydberg atoms and ion-Rydberg atom hybrid systems.
Using the ion microscope, we observed a novel molecular ion, where the bonding mechanism is based on the interaction between the ionic charge and an induced flipping dipole of a Rydberg atom [2]. Furthermore, we could measure the vibrational spectrum and spatially resolve the bond length and the angular alignment of the molecule. The excellent time resolution of the microscope enables probing of the interaction dynamics between the Rydberg atom and the ion.
[1] C. Veit, N. Zuber, O. A. Herrera-Sancho, V. S. V. Anasuri, T. Schmid, F. Meinert, R. Löw, and T. Pfau, Pulsed Ion Microscope to Probe Quantum Gases, Phys. Rev. X 11, 011036 (2021).
[2] N. Zuber, V. S. V. Anasuri, M. Berngruber, Y.-Q. Zou, F. Meinert, R. Löw, T. Pfau, Spatial imaging of a novel type of molecular ions, Nature 5, 453 (2022)"

Rydberg molecular ion and pulsed microscope team





Nicolas Zuber, Viraatt S. V. Anasuri, Moritz Berngruber
Yiquan Zou, Florian Meinert, Robert Löw

Christian Veit,
Oscar Herrera-Sanchez

erc European Research Council

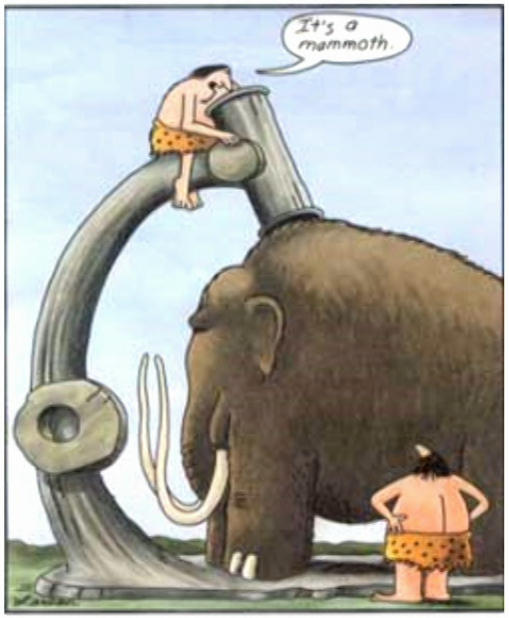
iRyd

Deutsche Forschungsgemeinschaft DFG

Universität Stuttgart
5. Physikalisches Institut

IQST Integrated Quantum Science and Technology

Carl Zeiss Stiftung



Early microscopes

Imaging the quantum world in real space

- A new pulsed ion microscope
- A new Rydberg molecule
- Spatial and temporal resolution of molecular vibrations
- Outlook

C. Veit et al. *Phys. Rev. X* **11**, 011036 (2021)

N. Zuber et al. *Nature* **5**, 453 (2022)

New pulsed ion microscope for ultracold atoms

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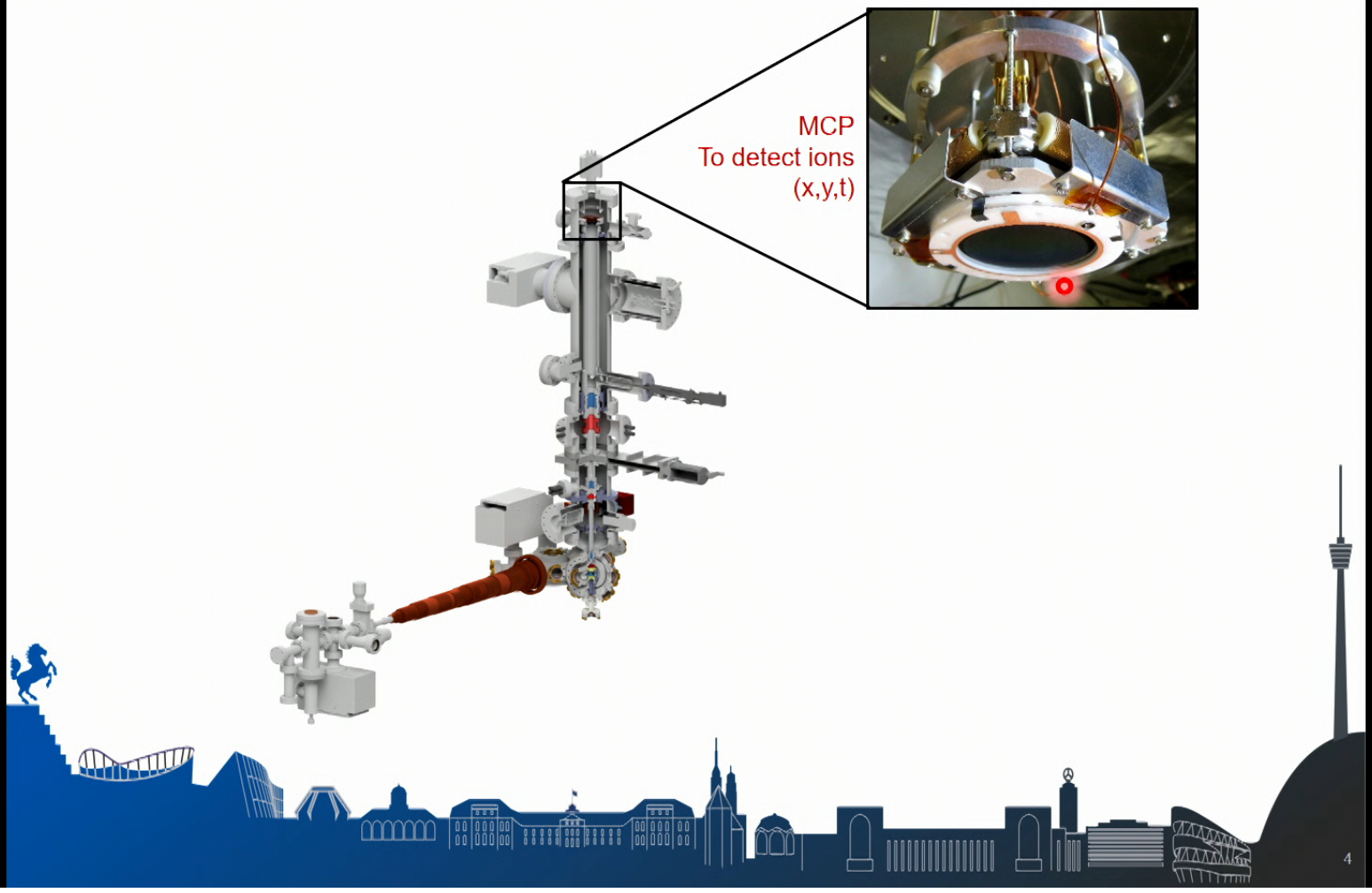

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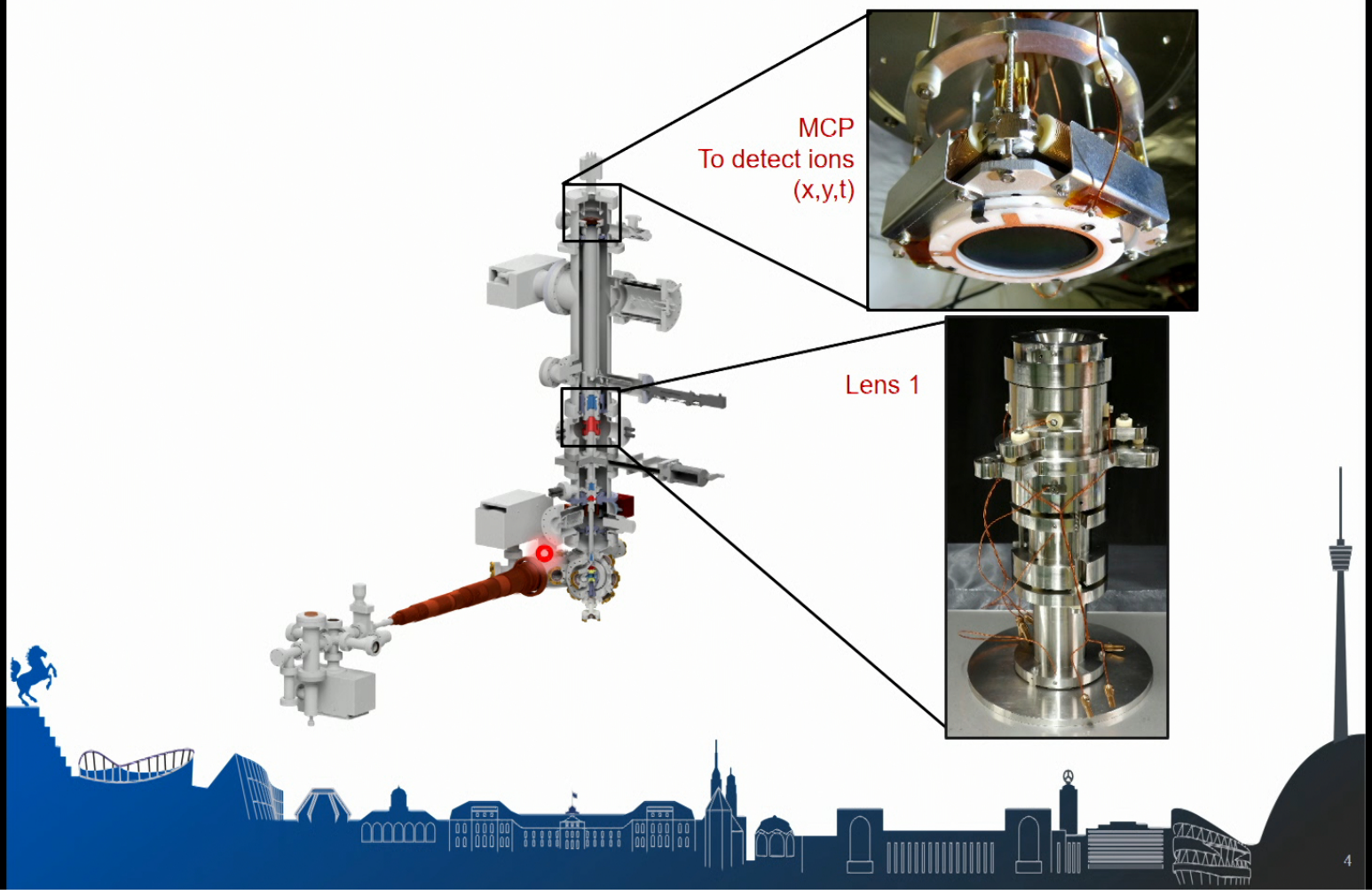

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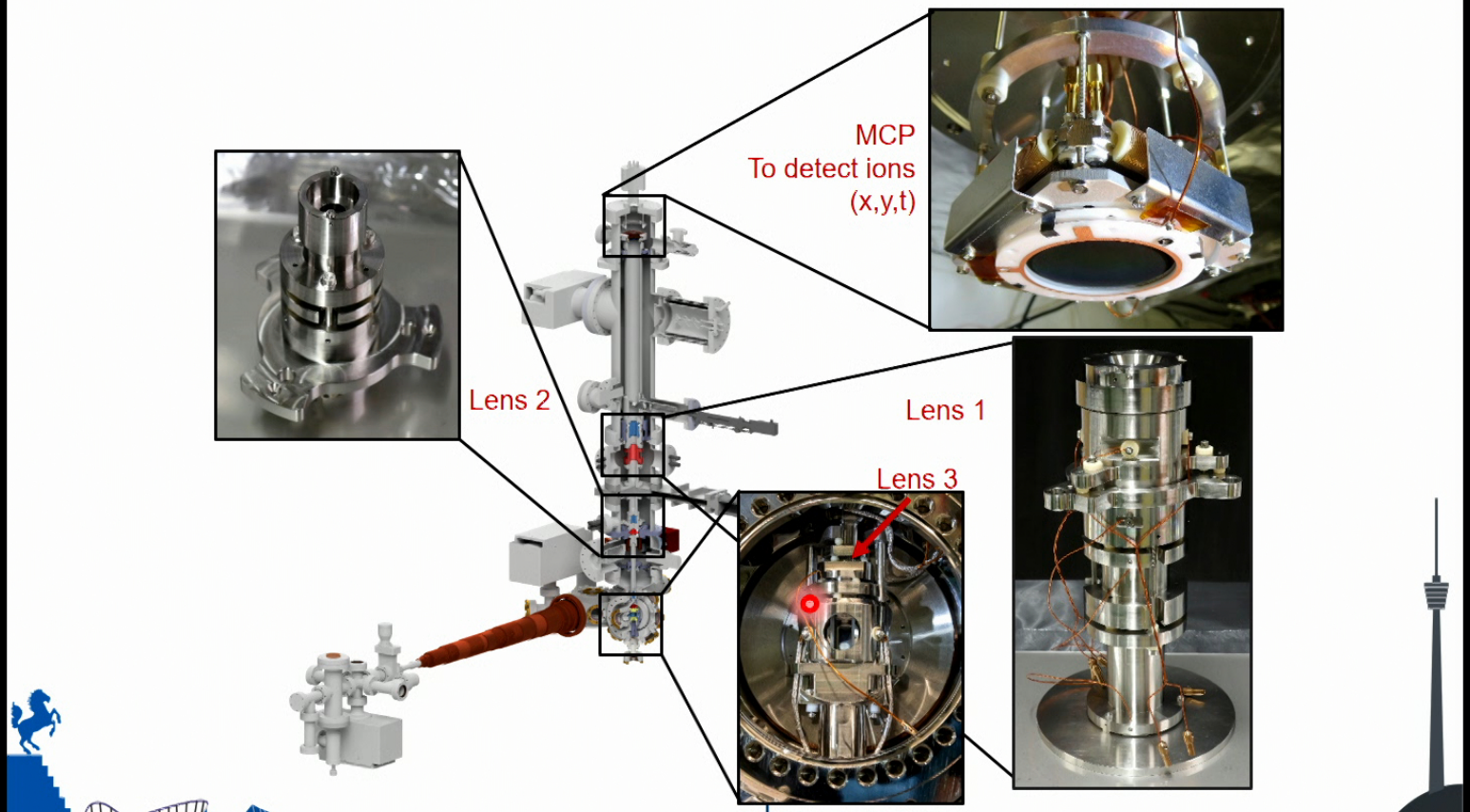

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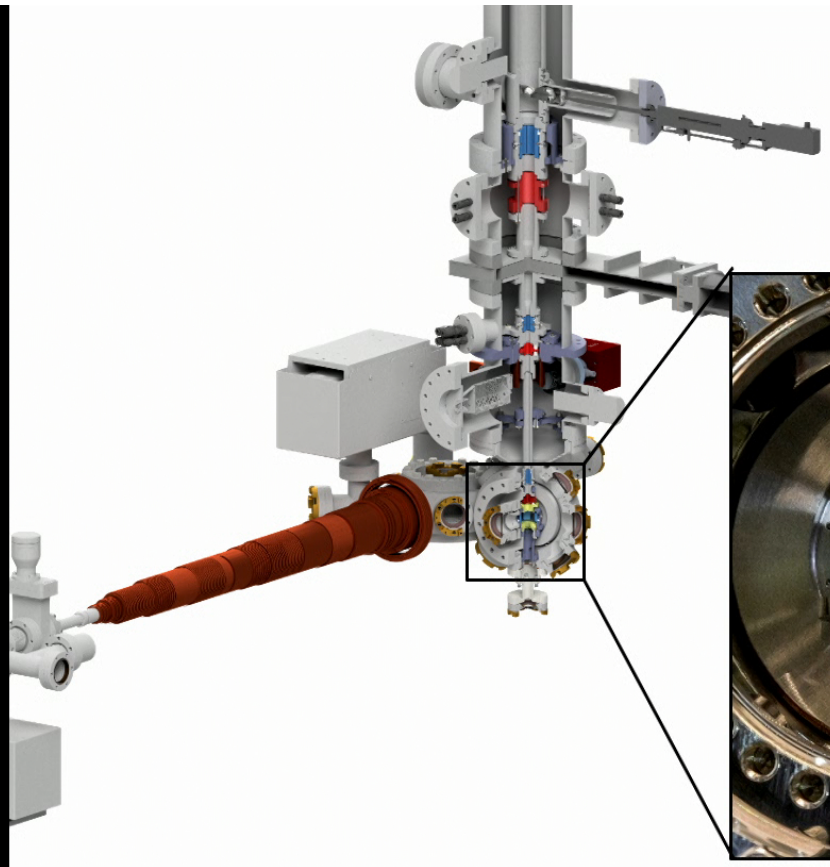
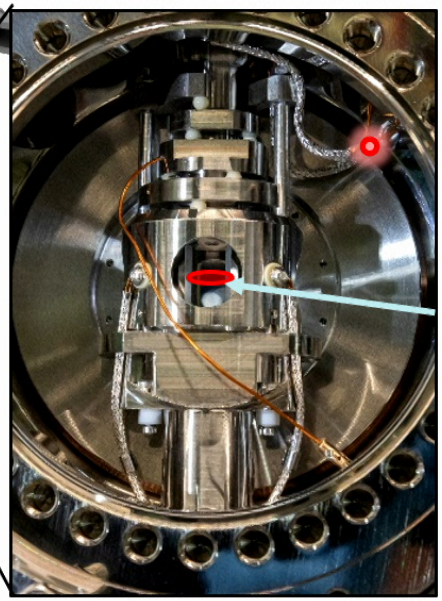
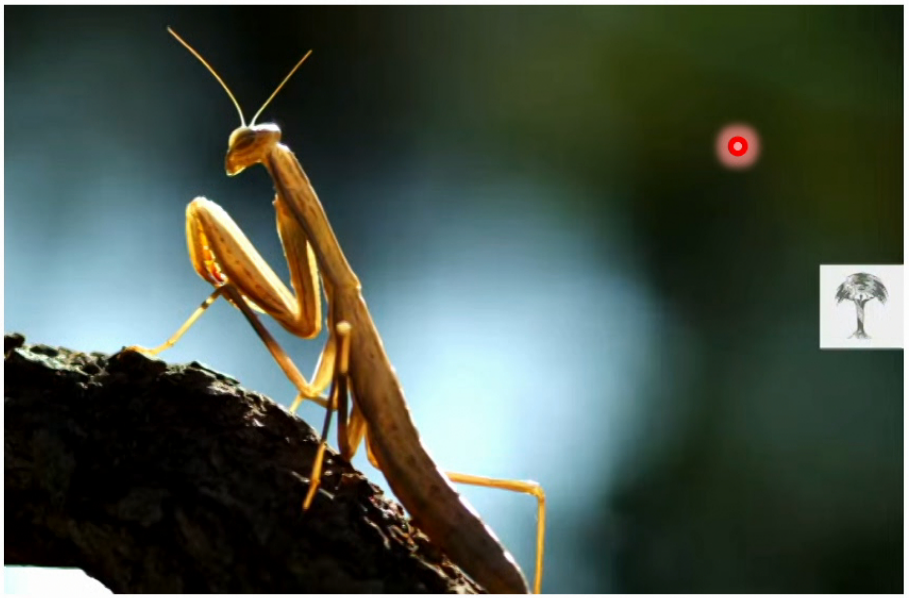
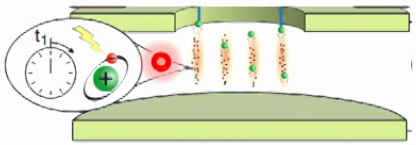




Cold atoms
in a tin can



New pulsed ion microscope for ultracold atoms



See also
Nat. Phys. **4**, 949 (2008, Ott)
Phys. Rev. Lett. **107**, 103001 (2011, Raithel)
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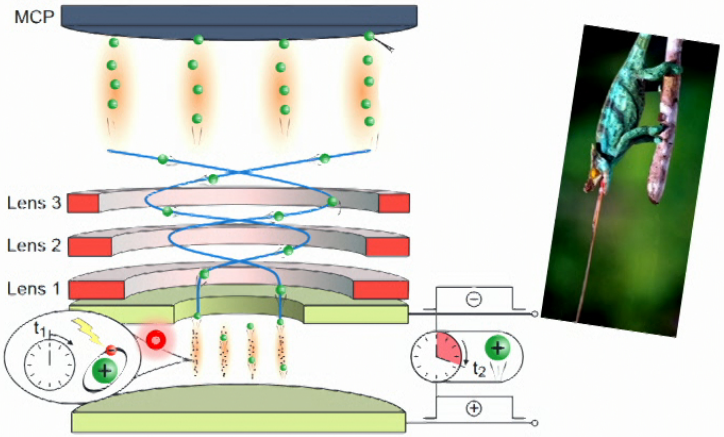
C. Veit et al. Phys. Rev. X **11**, 011036 (2021)

New pulsed ion microscope for ultracold atoms



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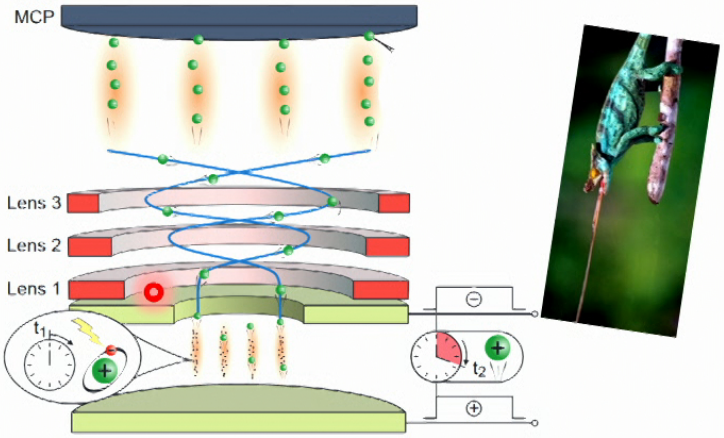

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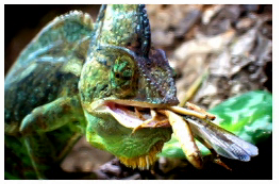
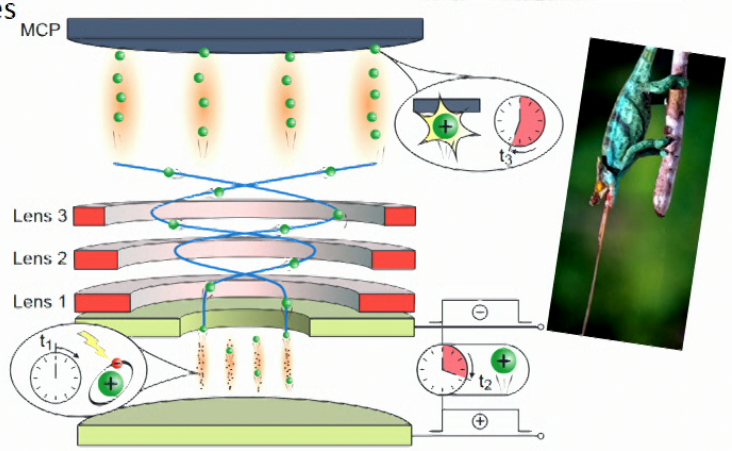
New pulsed ion microscope for ultracold atoms

Benefits of ion microscope to observe and control single atoms

- imaging of ionized ground state atoms, Rydbergs & (ultracold) ionic impurities
- High time resolution (<100 nsec)
- High spatial resolution (<200nm)
- 3D-imaging (large depth of field)
- Very good E-Field control:
  free cold ions
- ...





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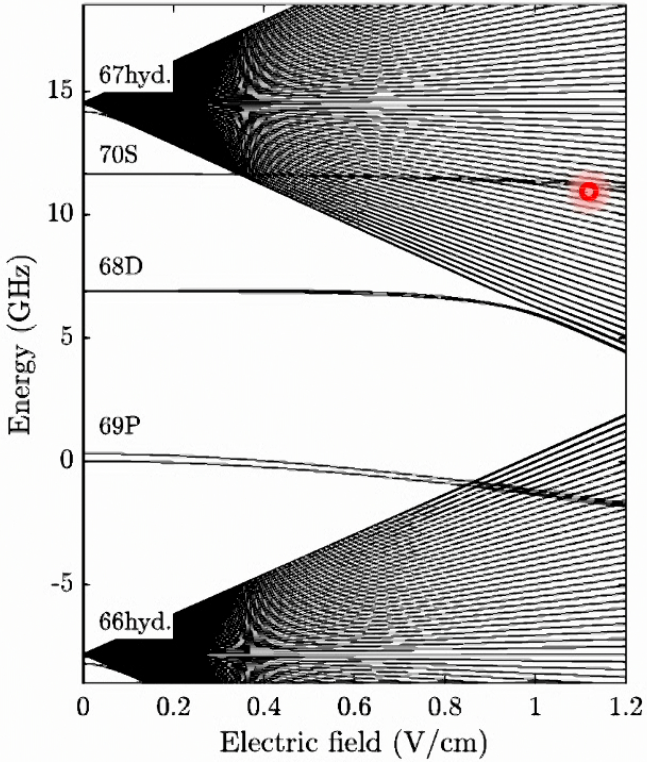
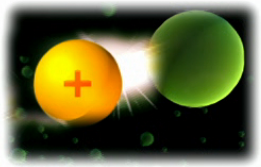
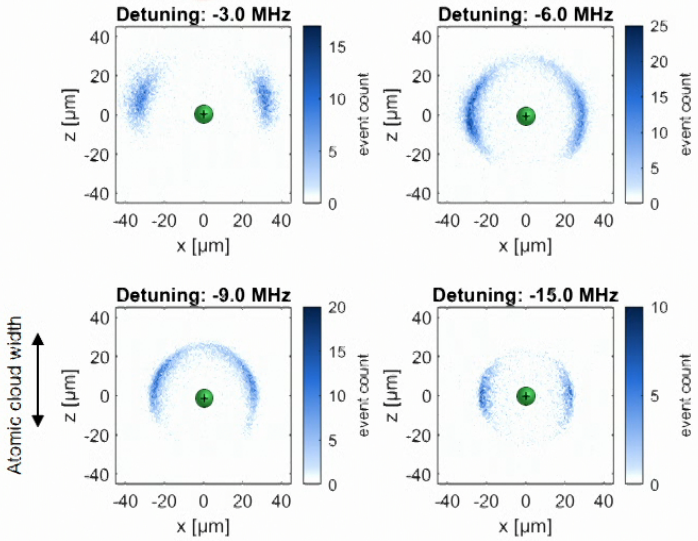
Reminder: Stark map of Rubidium



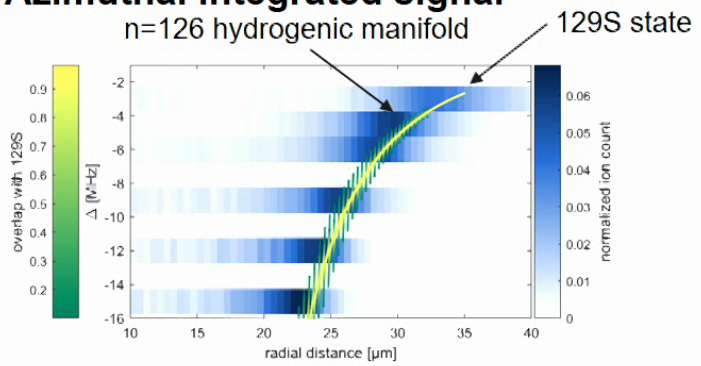
Probing the ion – Rydberg atom potential



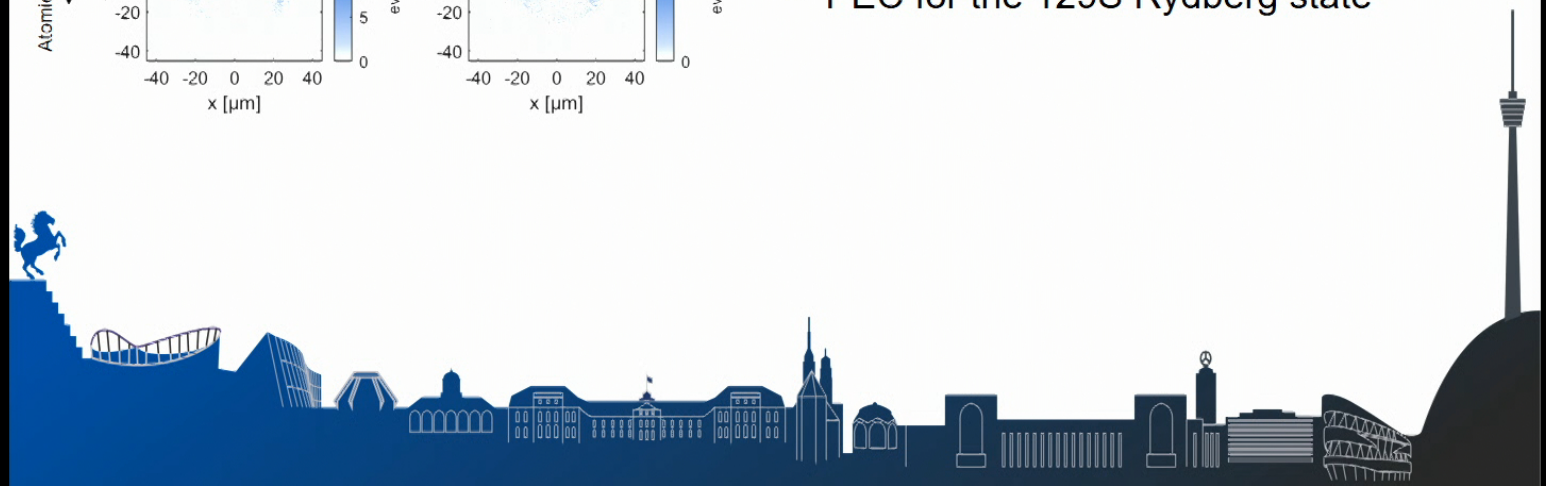
2D images: facilitated excitation



Azimuthal integrated signal



- PEC for the 129S Rydberg state



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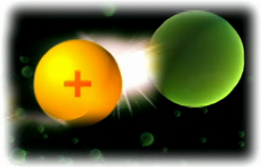
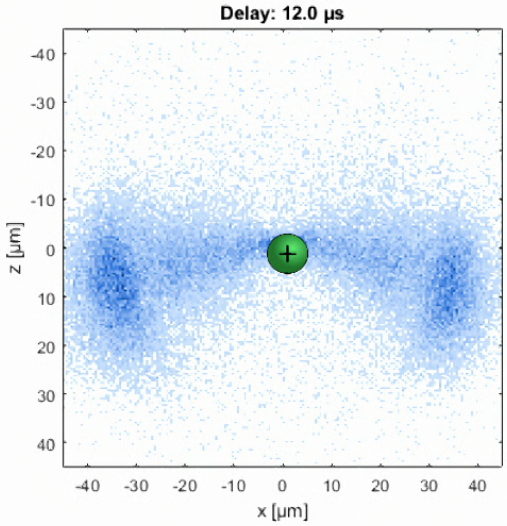




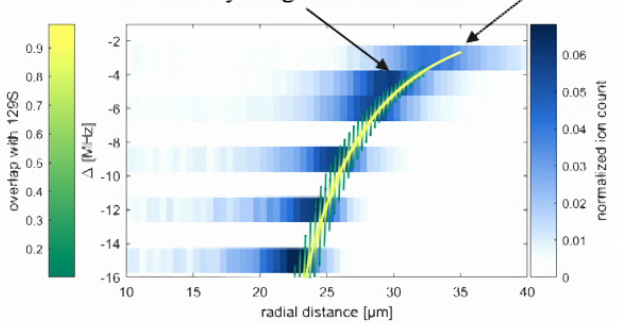
Detuning -3 MHz

Preliminary data

Azimuthal integrated signal

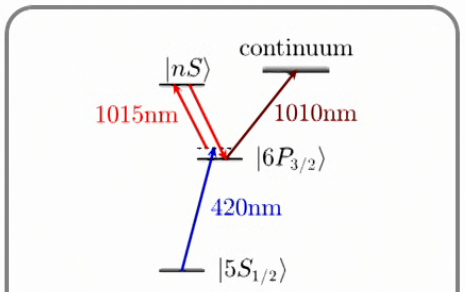
n=126 hydrogenic manifold

129S state

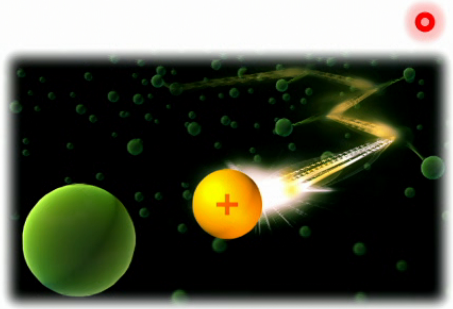


- PEC for the 129S Rydberg state

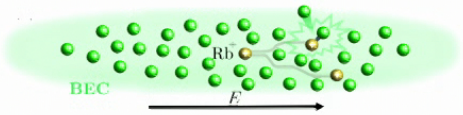
Make a single ion and study transport



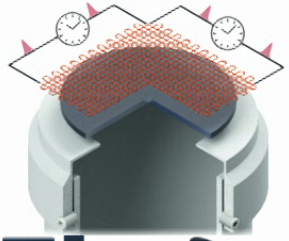
Low-energy ions from single Rydberg atoms



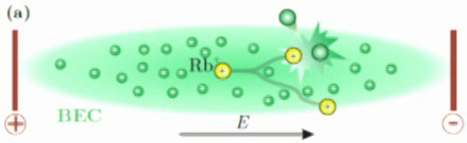
Ionic transport through a BEC



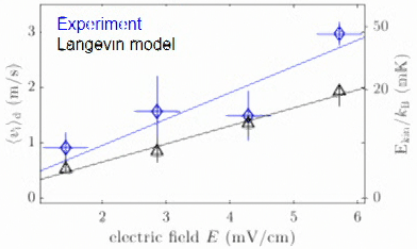
A microscope for ions

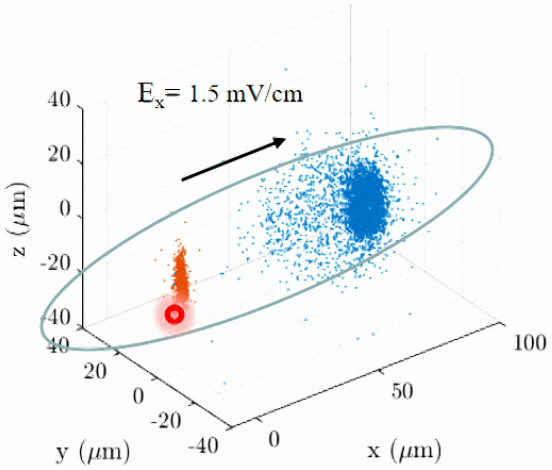


Motivation: See charge transport in a cloud of atoms



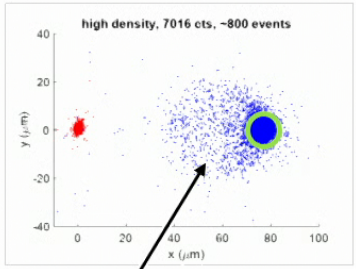
Diffusive transport in BEC



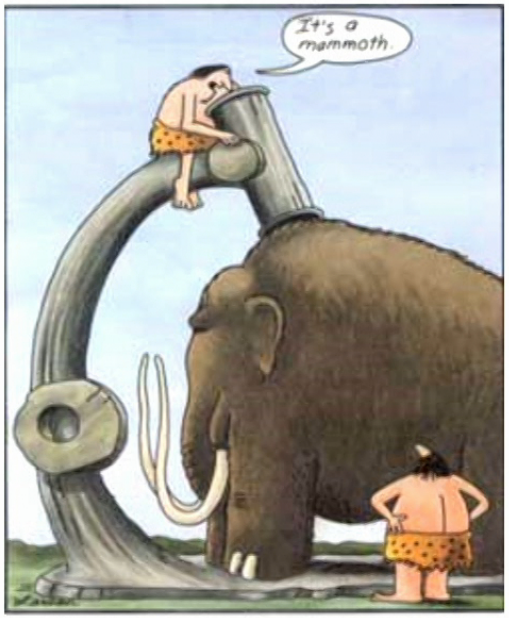


T. Dieterle et al., PRL. **126**, 033401 (2021)
Transport of a Single Cold Ion Immersed in a Bose-Einstein Condensate

See Viewpoint in Physics



Scattering halo of a single Langevin collision under the microscope (30 μs TOF)



Early microscopes

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- A new Rydberg molecule
- Spatial and temporal resolution of molecular vibrations
- Outlook

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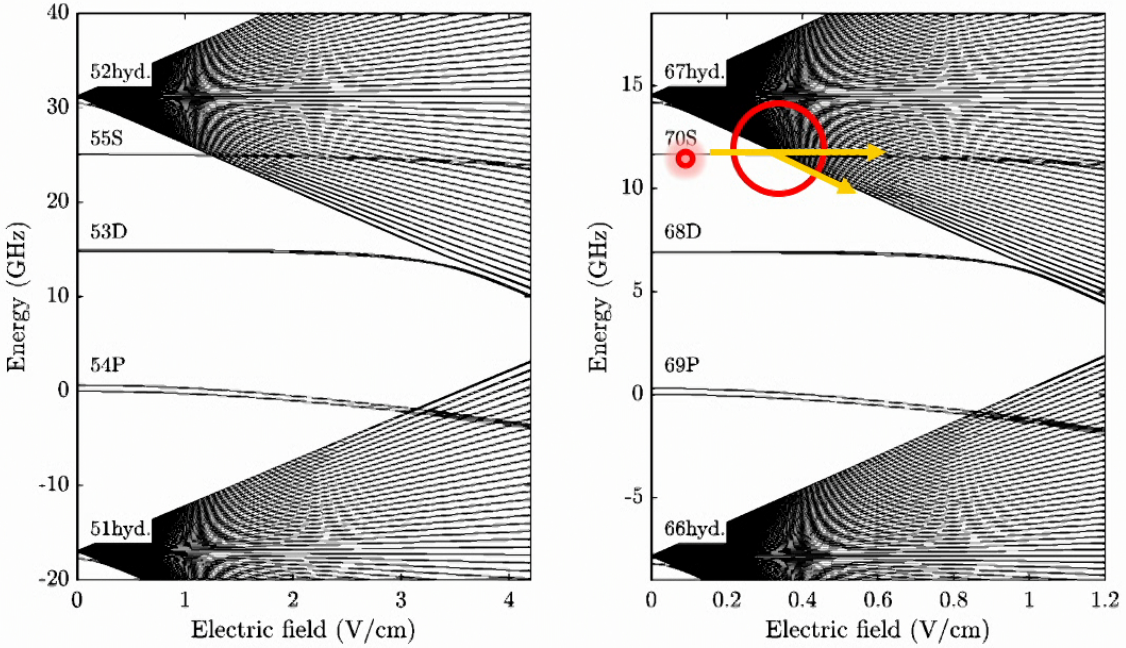
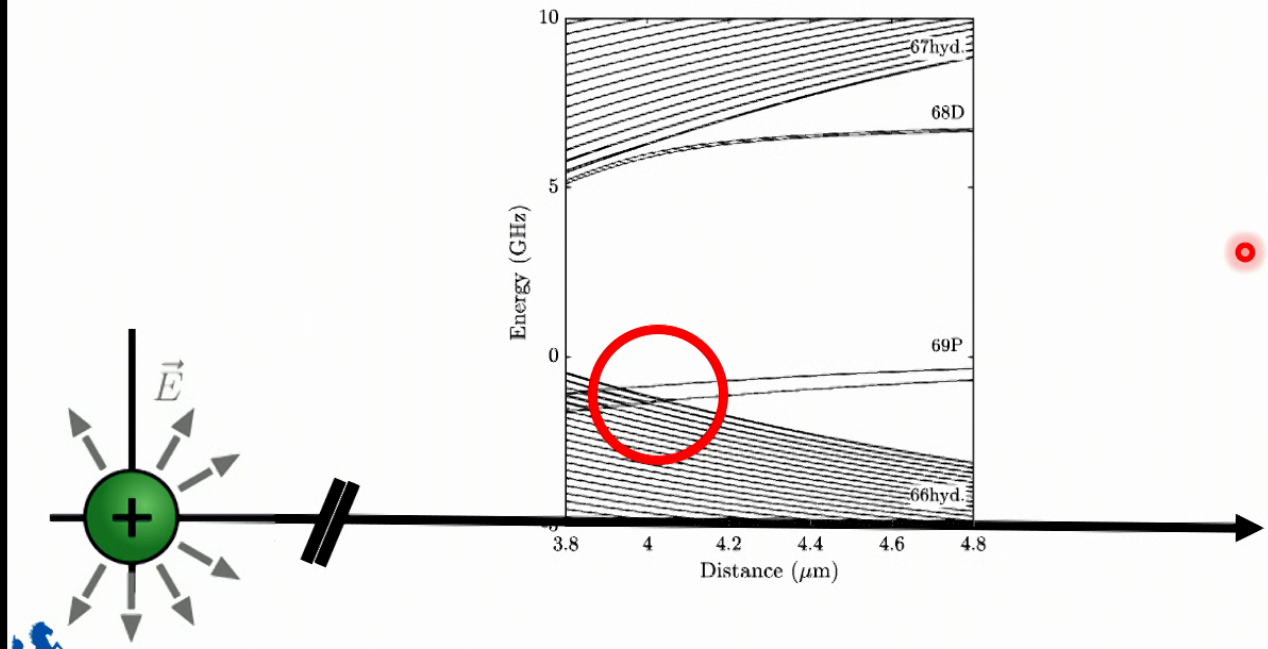
N. Zuber et al. *Nature* **5**, 453 (2022)

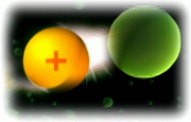
Stark Effect: Rydberg atom in Electric field



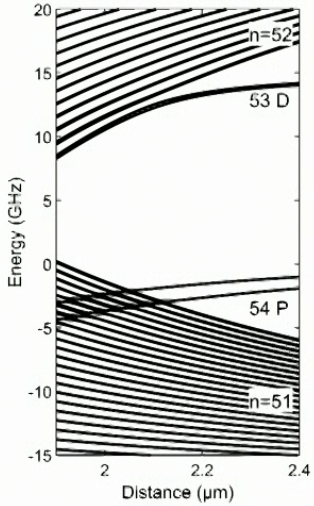
Rydberg Atom in the field of an ion



Ion & Rydberg atom interaction



- Electric field of the ion polarizes the Rydberg atom
-



Proposal:

[1] A. Duspayev, et al Phys. Rev. Research **3**, 023114 (2021).
[2] M. Deiß, S. Haze, and J. Hecker Denschlag,, Atoms **9**, 2 (2021).

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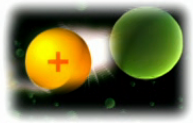
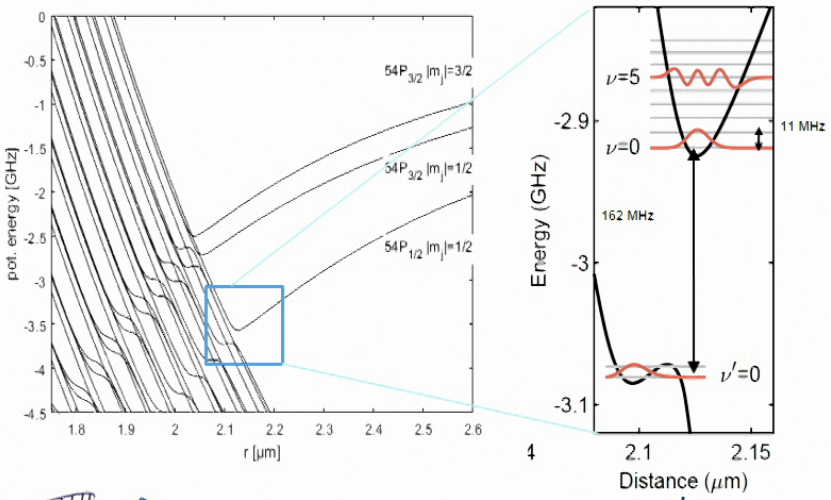
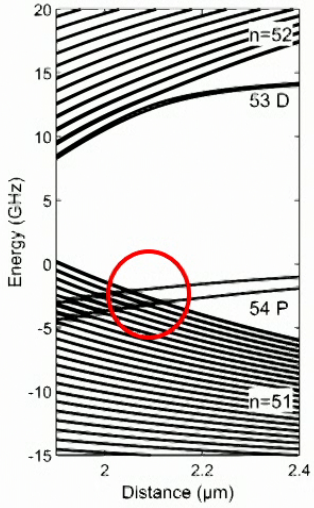


- Electric field of the ion polarizes the Rydberg atom
- inhomogeneous field:

  -> taking multipole terms up to the 6th order into account [1,2]

$$V_I = \frac{-e^2}{4\pi\epsilon_0} \sum_{l=1}^{l_{max}} \sqrt{\frac{4\pi}{2l+1}} \frac{r_e^l}{r^{l+1}} \Upsilon_{l0}(\theta_e, \phi_e)$$





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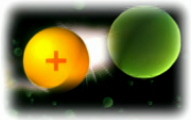

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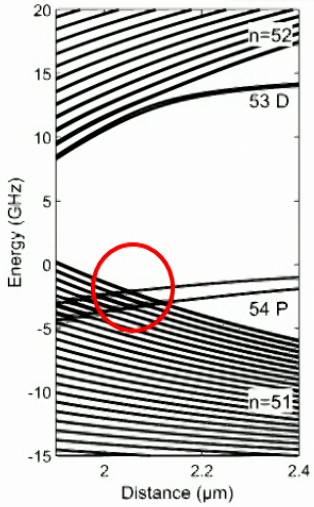

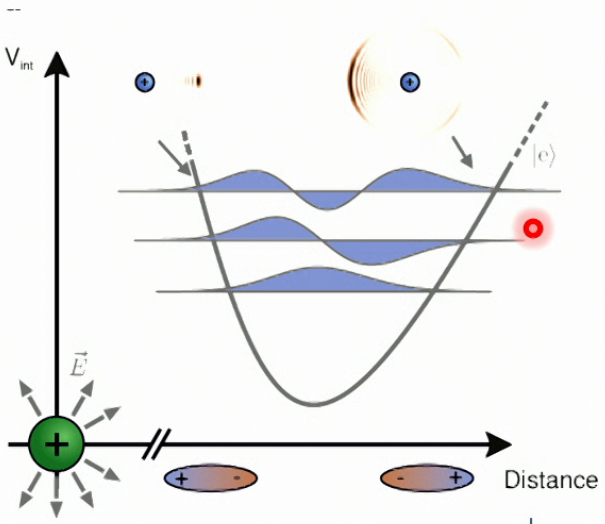

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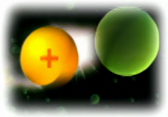
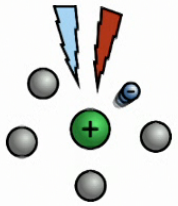
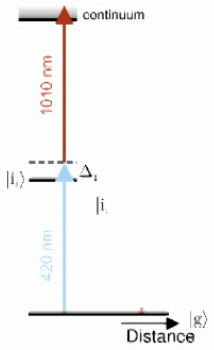


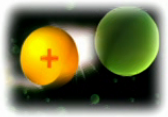
Making a new molecular ion: Bound ion & Rydberg atom

Creating cold ion
in field free space



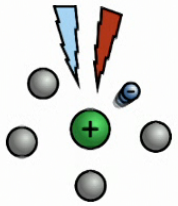
- 1μs two color ionization pulse of rubidium
- Ion excess energy $k_B \times few$ μK





Making a new molecular ion: Bound ion & Rydberg atom

Creating cold ion in field free space



- 1μs two color ionization pulse of rubidium
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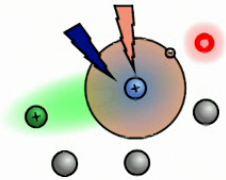
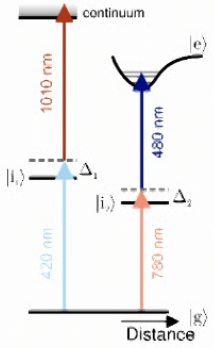
Rydberg excitation

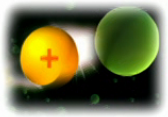
wait



- 1μs two color Rydberg excitation at detuning Δ

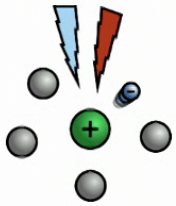


18



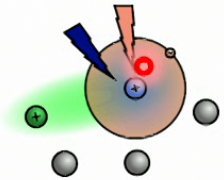
Making a new molecular ion: Bound ion & Rydberg atom

Creating cold ion in field free space

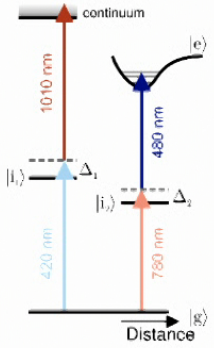


- 1μs two color ionization pulse of rubidium
- Ion excess energy $k_B \times few$ μK

Rydberg excitation

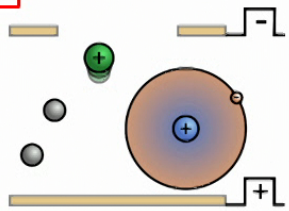


- 1μs two color Rydberg excitation at detuning Δ



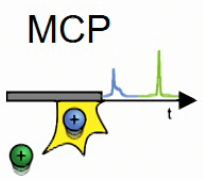
wait

Separation & extraction



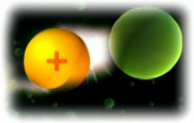
- Low field separation pulse
- Ionize the Rydberg atom in high field & extract both ions

Imaging & detection



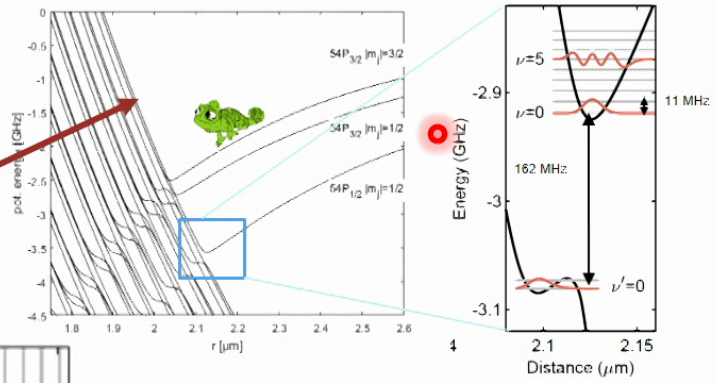
- Image with ion microscope
- Extract ion & Rydberg atom position

A single ion bound to a Rydberg atom: a new molecular ion

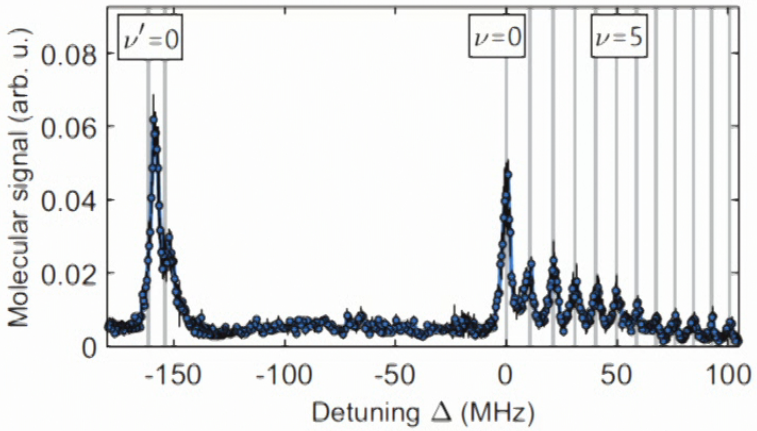


- PEC for $54P_{\frac{1}{2}}, |m_j| = \frac{1}{2}$

inner wall: high l states

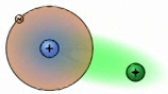


spectroscopy



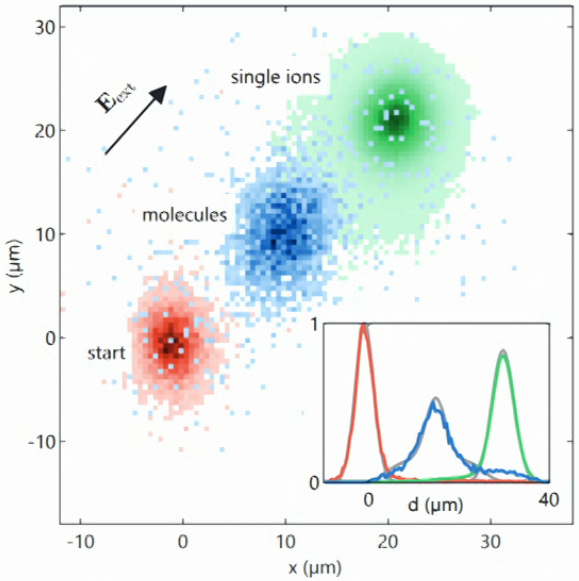
N. Zuber et al. *Nature* **5**, 453 (2022)

Proposal:
[1] A. Duspayev, et al., Phys. Rev. Research **3**, 023114 (2021).
[2] M. Deiß, S. Haze, and J. Hecker Denschlag, Atoms **9**, 2 (2021).



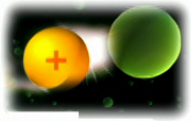
Mass spectroscopy in real space

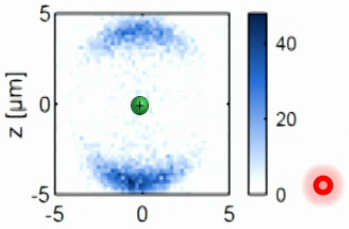
- Create molecules and ions at initial spot
- Apply small electric field $\vec{E}_{\mathrm{ext}} \approx 2.3\,{}^{\mathrm{mV}}/_{\mathrm{cm}}$
- Molecules travel half the distance in the same time due to doubled mass
- Most molecules stay bound



N. Zuber et al. *Nature* **5**, 453 (2022)

A single ion bound to a Rydberg atom: molecular alignment in real space





ent

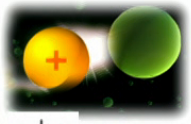
Measured bond length: 4.3 μm (theory: 4.1 μm)

In situ images of molecules for the 69P state

Z
X

N. Zuber et al. *Nature* **5**, 453 (2022)

A single ion bound to a Rydberg atom: molecular alignment in real space

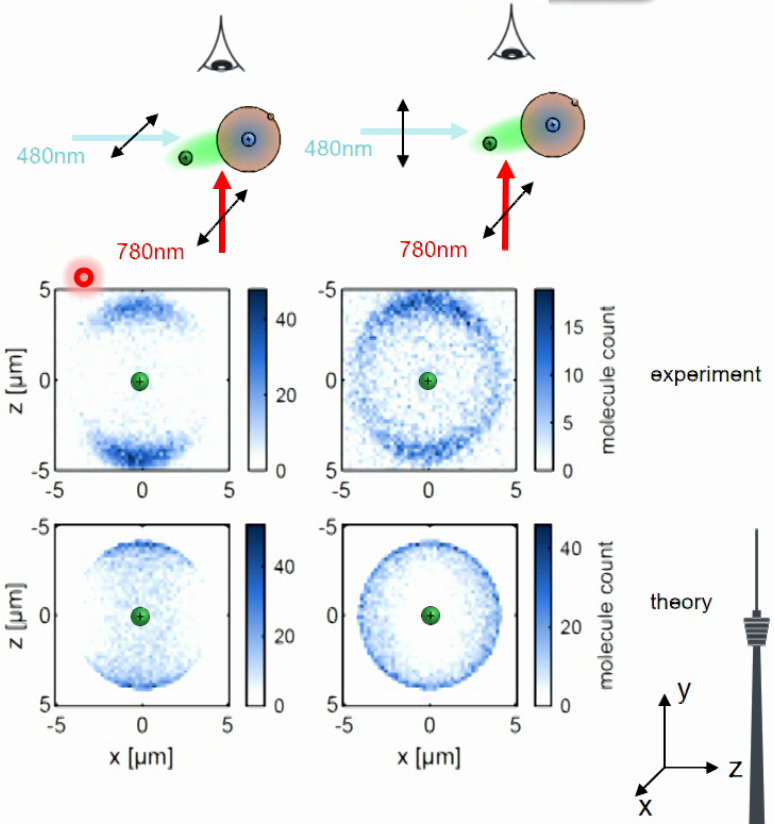


- Excitation probability depends on light polarization and molecular orientation
- $P_{g\to f} \propto |\sum_i \langle f|\vec{\epsilon}_2\vec{d}|i\rangle\langle i\left|\vec{\epsilon}_1\vec{d}\right|g\rangle|^2$

  $|g\rangle = |5S_{\frac{1}{2}}, F=2\rangle$ ground state

  $|i\rangle = |5P_{\frac{3}{2}}, F=3\rangle$ intermediate state

  $|f\rangle$ Rydberg state



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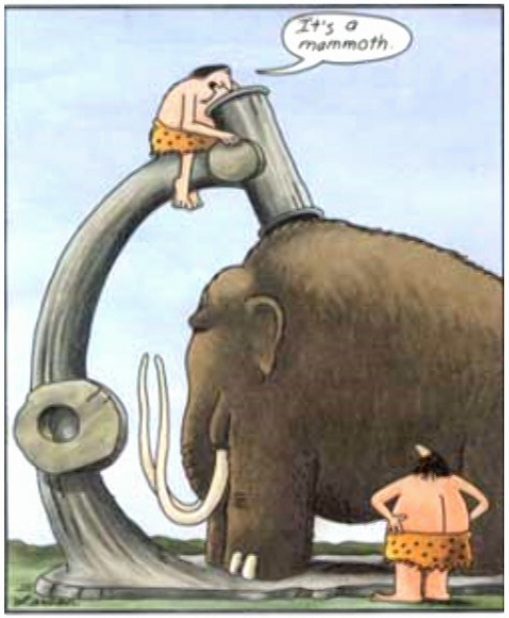
N. Zuber et al. *Nature* **5**, 453 (2022)



Early microscopes

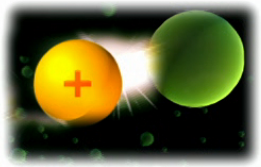
Imaging the quantum world in real space

- A new pulsed ion microscope
- A new Rydberg molecule
- Spatial and temporal resolution of molecular vibrations
- Outlook

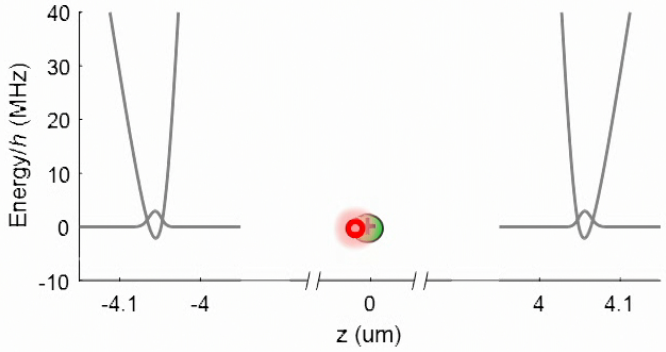
C. Veit et al. *Phys. Rev. X* **11**, 011036 (2021)

N. Zuber et al. *Nature* **5**, 453 (2022)

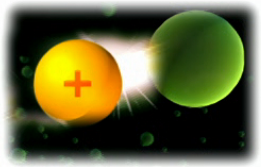
E-field induced wave packet preparation:



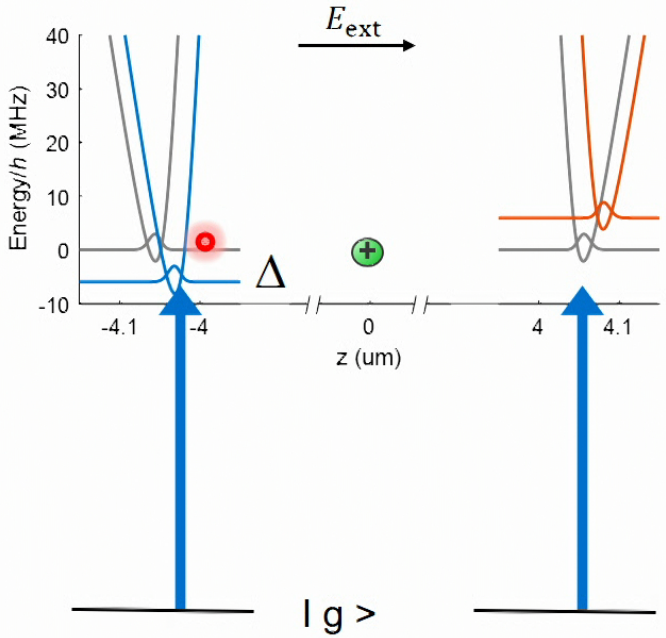
Molecular potential

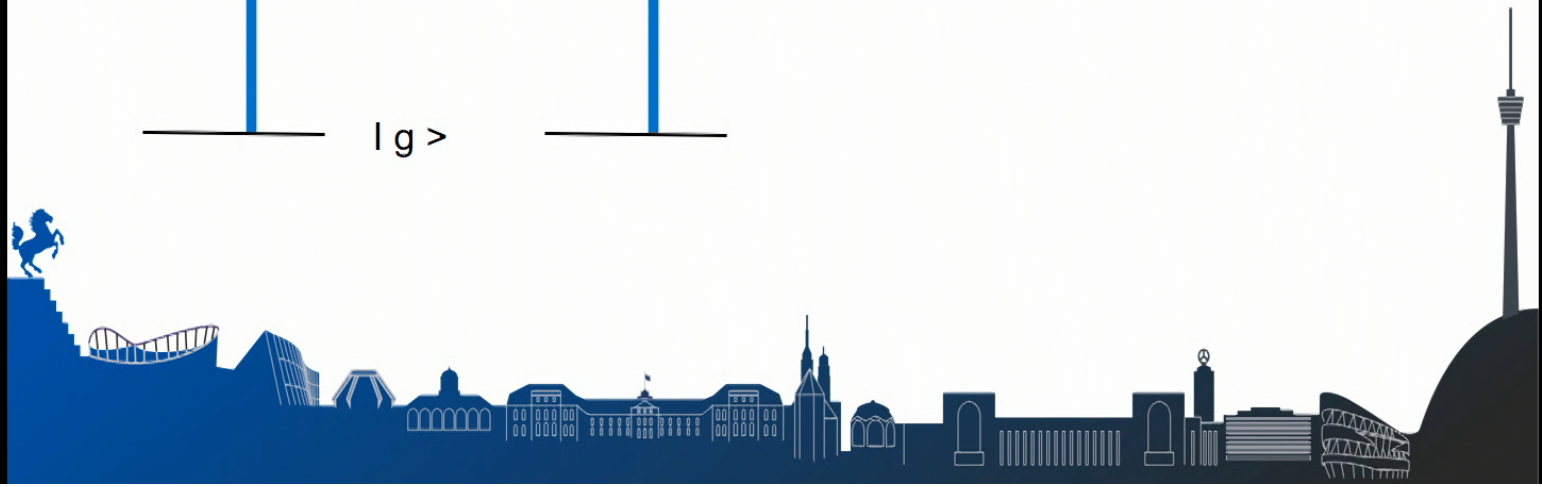


E-field induced wave packet preparation:

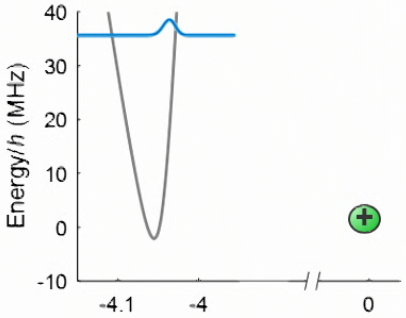


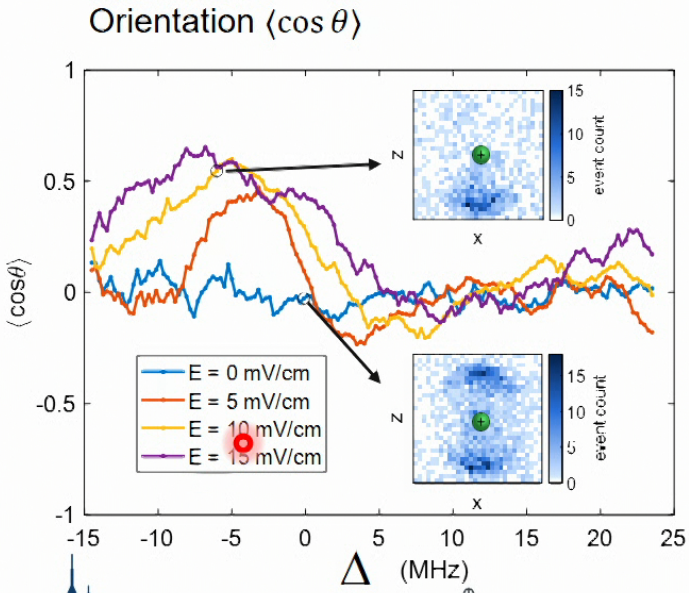
Molecular potential





Vibrational wave packet dynamics:





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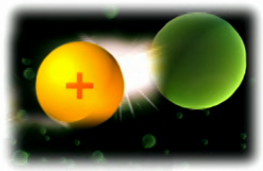
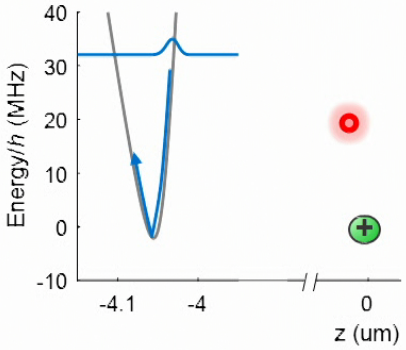
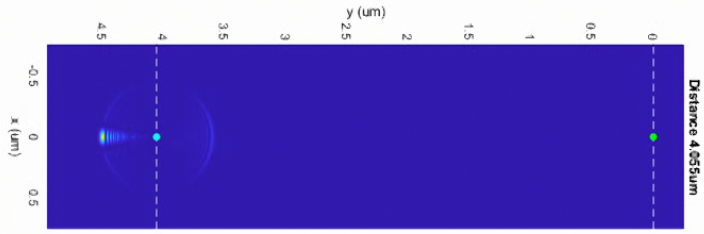
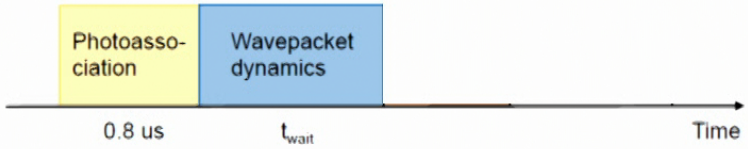
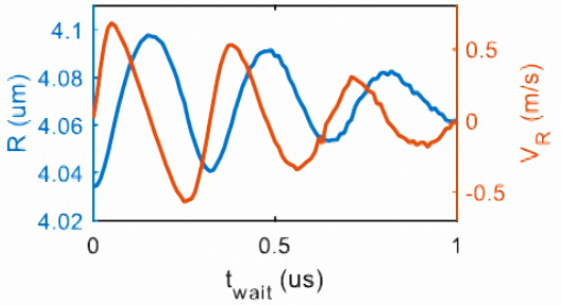




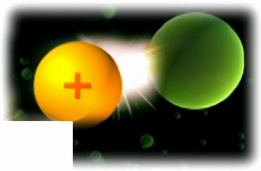


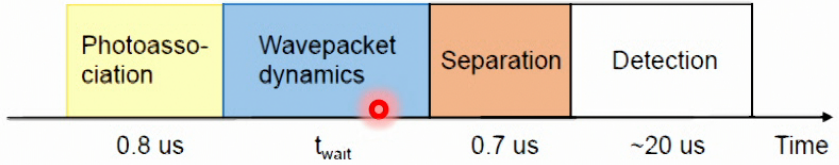


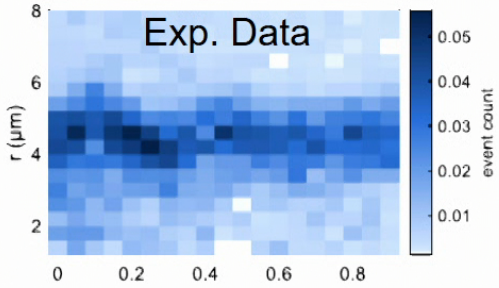
Expected phase space dynamics

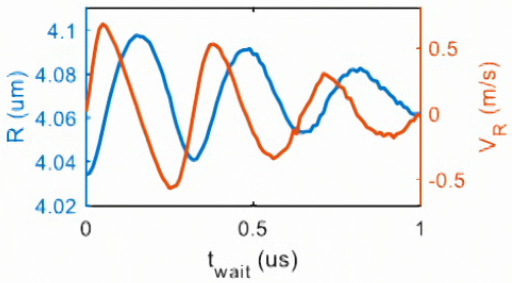


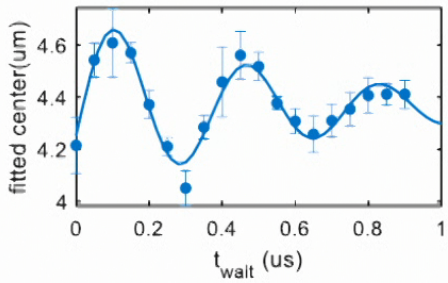
Vibrational wave packet dynamics:

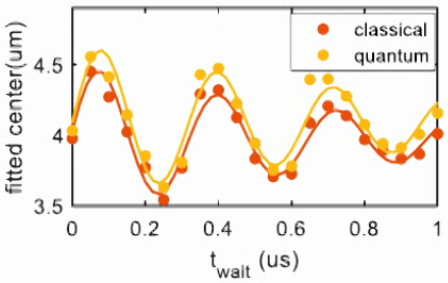








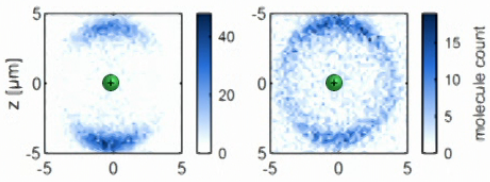




Experiment:
f = 2.8 MHz

Classical simulation:
f = 3.2 MHz

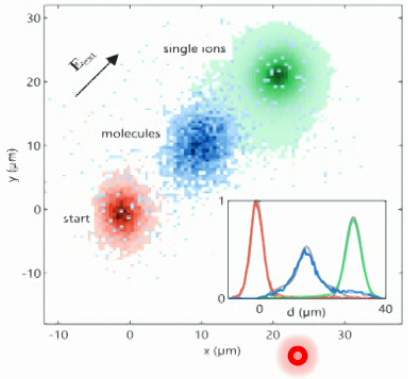
Summary

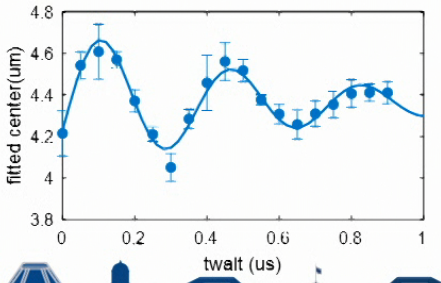


New Atom – Ion bound state

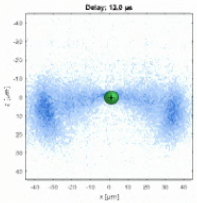
fast vibrational dynamics

Observed in real space



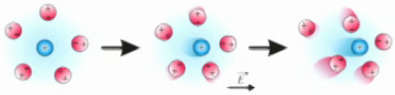


Outlook

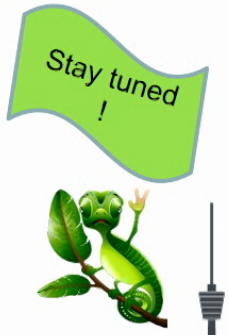


Rydberg Chemistry:
Atom - Ion dynamics in real space

- ultracold ionic impurities in a degenerate quantum gas, polarons, transport etc.



- Friedel oscillations in e.g. ^{6}Li
- observation of ion-atom collisions in the quantum regime PRL**120**, 153401 (2018)
- Rydberg-Rydberg correlations in 3D
- Molecular ions: Trimers, Dynamics….



Our team in Stuttgart:

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We are hiring PhD students and Postdocs!