

**Universität  
Stuttgart**



# Rydberg molecules

**Pisa**

**Sept, 18<sup>th</sup> - 2012**

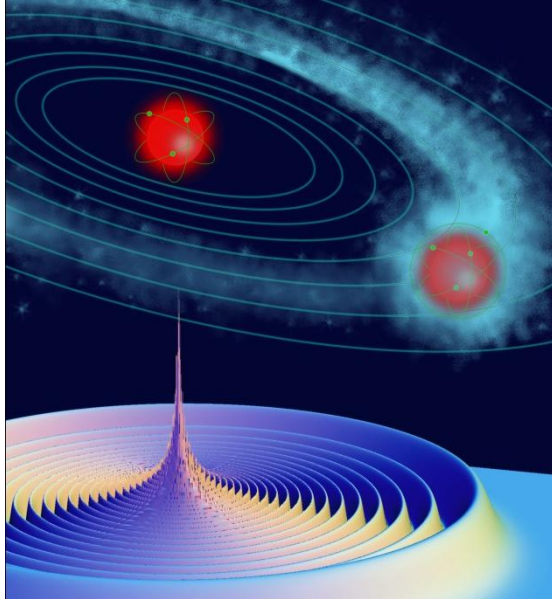
**Vera Bendkowsky, Björn Butscher, Johannes Nipper, Jonathan  
Balewski, Robert Löw & Tilman Pfau**

**H Sadeghpour, JM Rost, T Pohl, S Rittenhouse, W Li, J Shaffer**





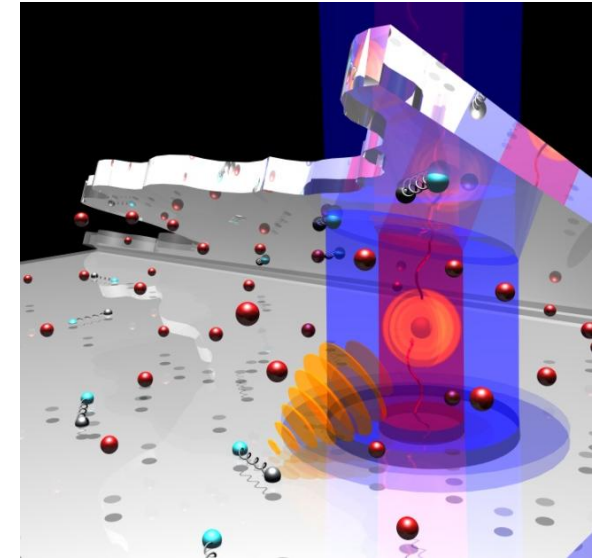
# Research topics



Rydberg molecules



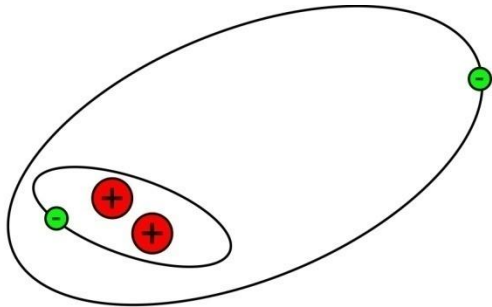
Frozen Rydberg gases



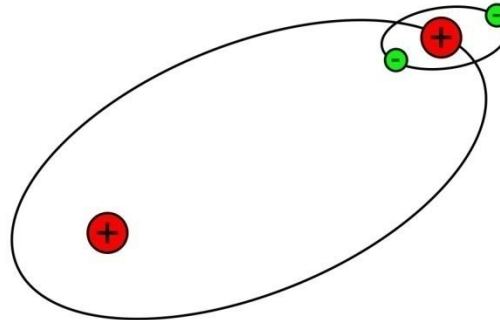
$\mu$ -cells



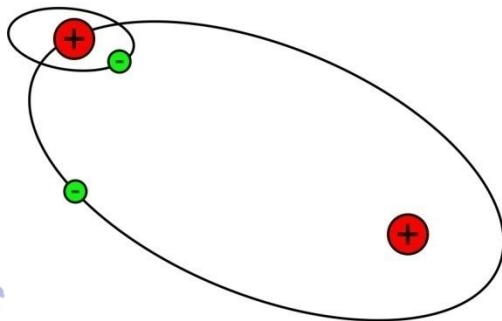
# Rydberg molecules



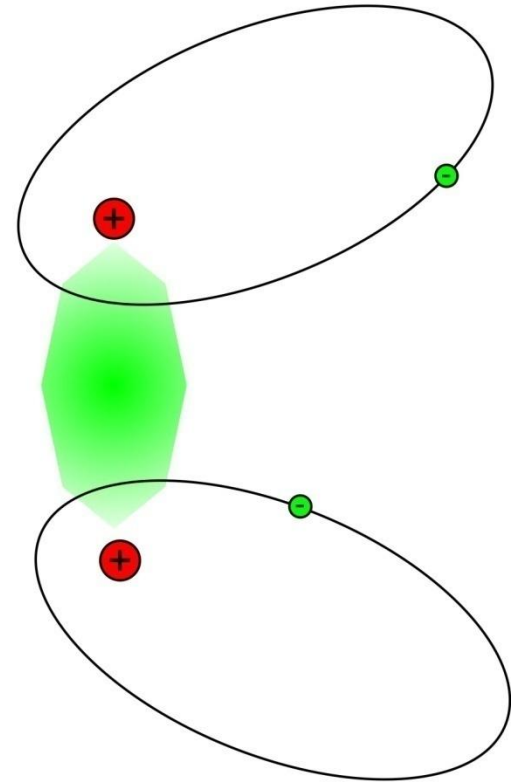
Ordinary Rydberg molecules



Heavy Rydberg



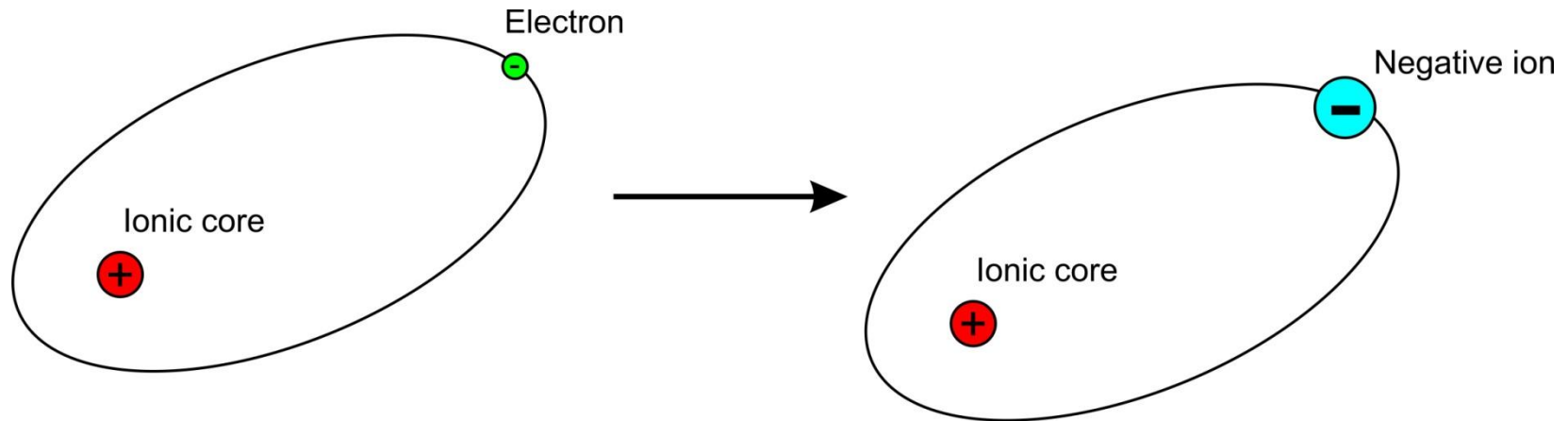
Trilobite Rydberg molecules



Rydberg-Rydberg molecules



# Heavy Rydberg



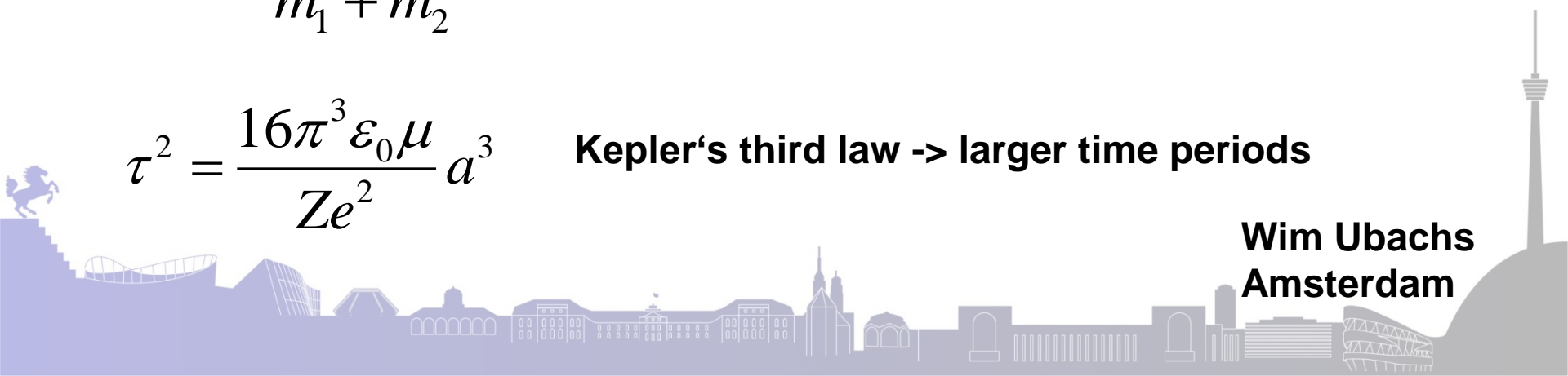
$$\mu = \frac{m_1 m_2}{m_1 + m_2}$$

Larger reduced mass

$$\tau^2 = \frac{16\pi^3 \epsilon_0 \mu}{Z e^2} a^3$$

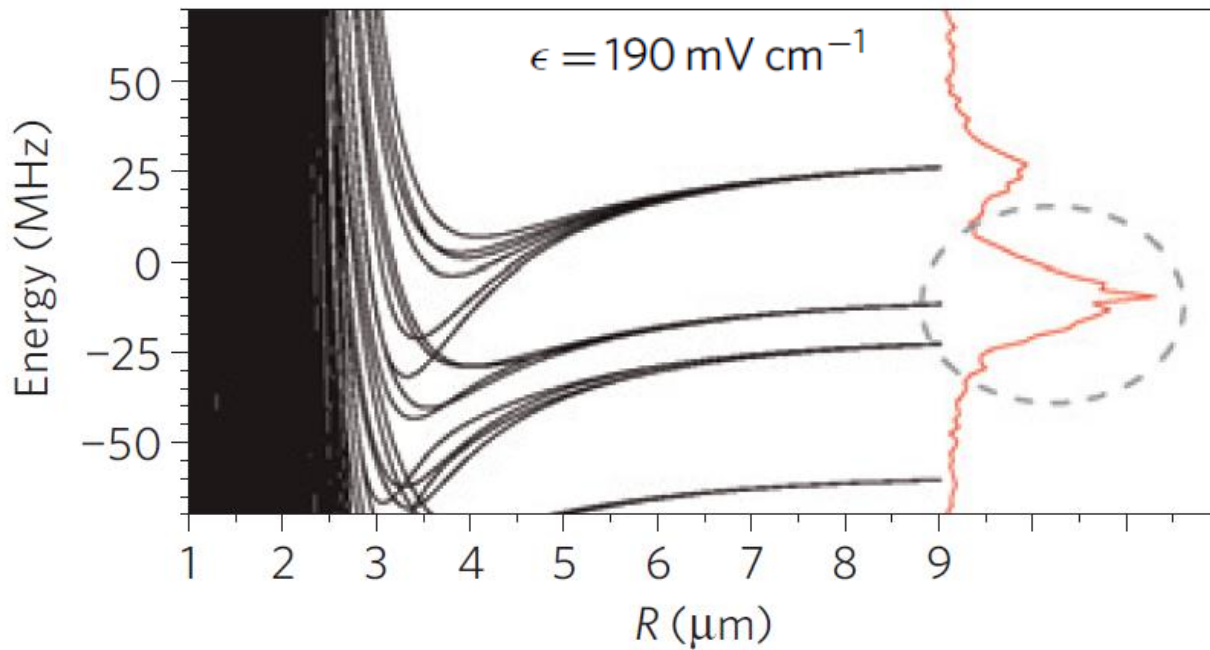
Kepler's third law -> larger time periods

Wim Ubachs  
Amsterdam





# Observation of Cs Rydberg atom macrodimers



$65D_{5/2}67D_{5/2}$

$|m_{j1}| = 1/2 |m_{j2}| = 1/2$

$|m_{j1}| = 3/2 |m_{j2}| = 1/2$

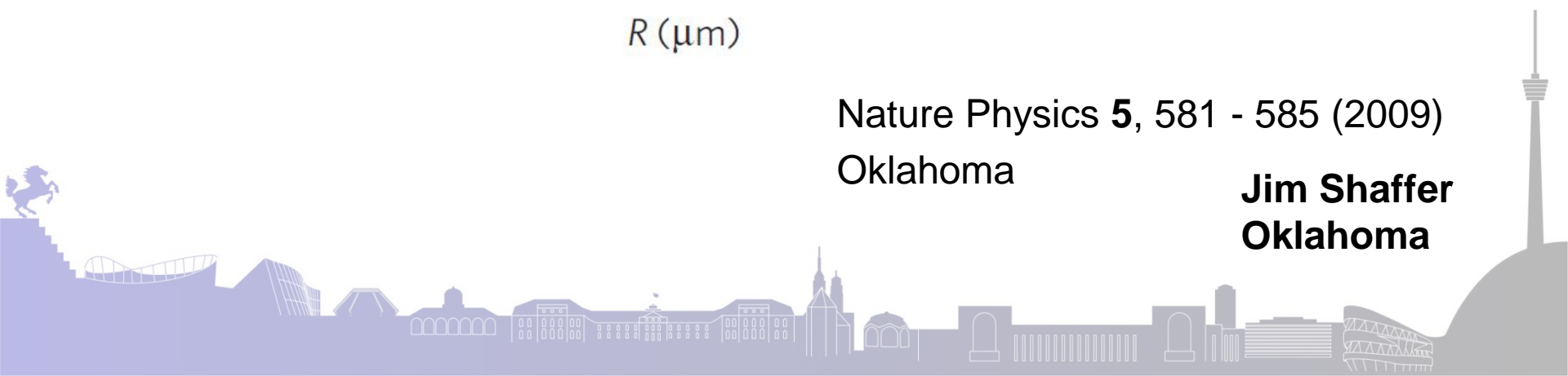
$|m_{j1}| = 1/2 |m_{j2}| = 3/2$

$|m_{j1}| = 3/2 |m_{j2}| = 3/2$

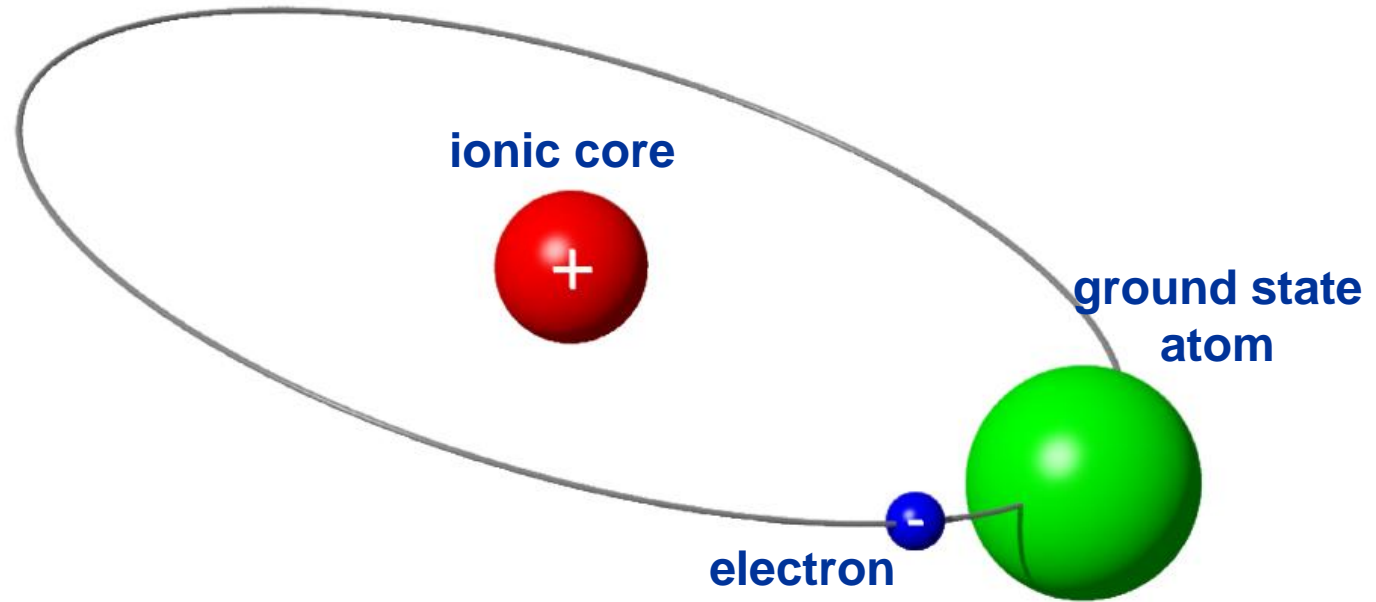
Nature Physics **5**, 581 - 585 (2009)

Oklahoma

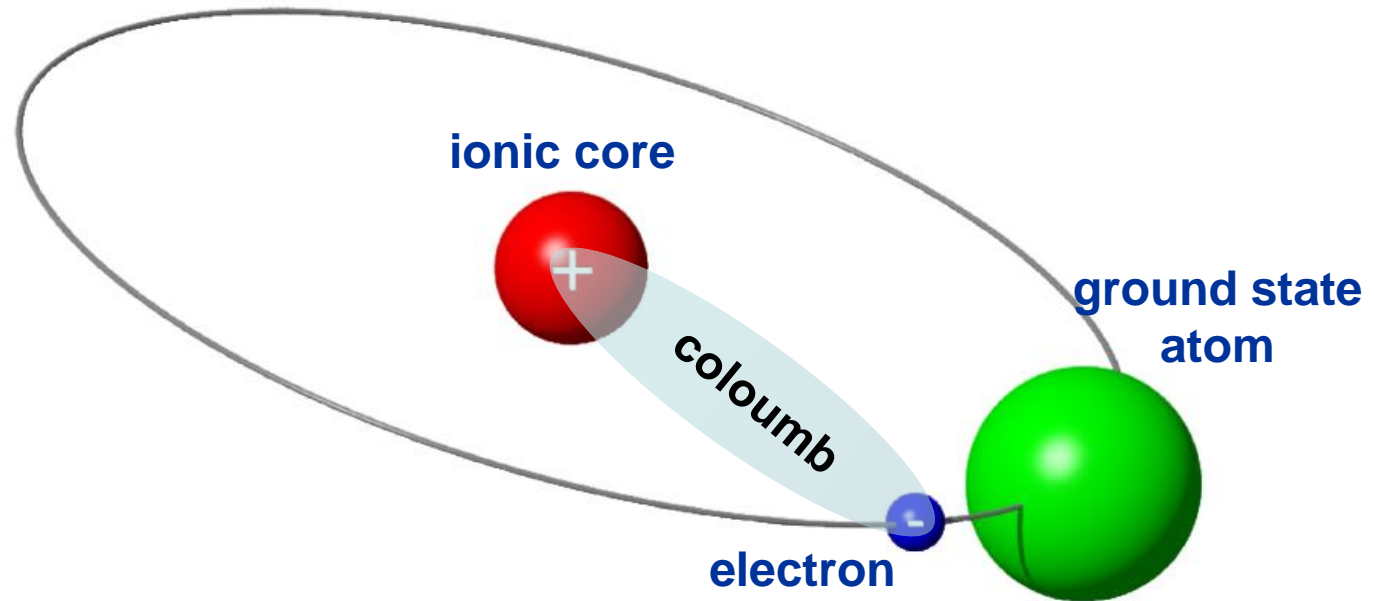
**Jim Shaffer**  
**Oklahoma**



# Rydberg molecules

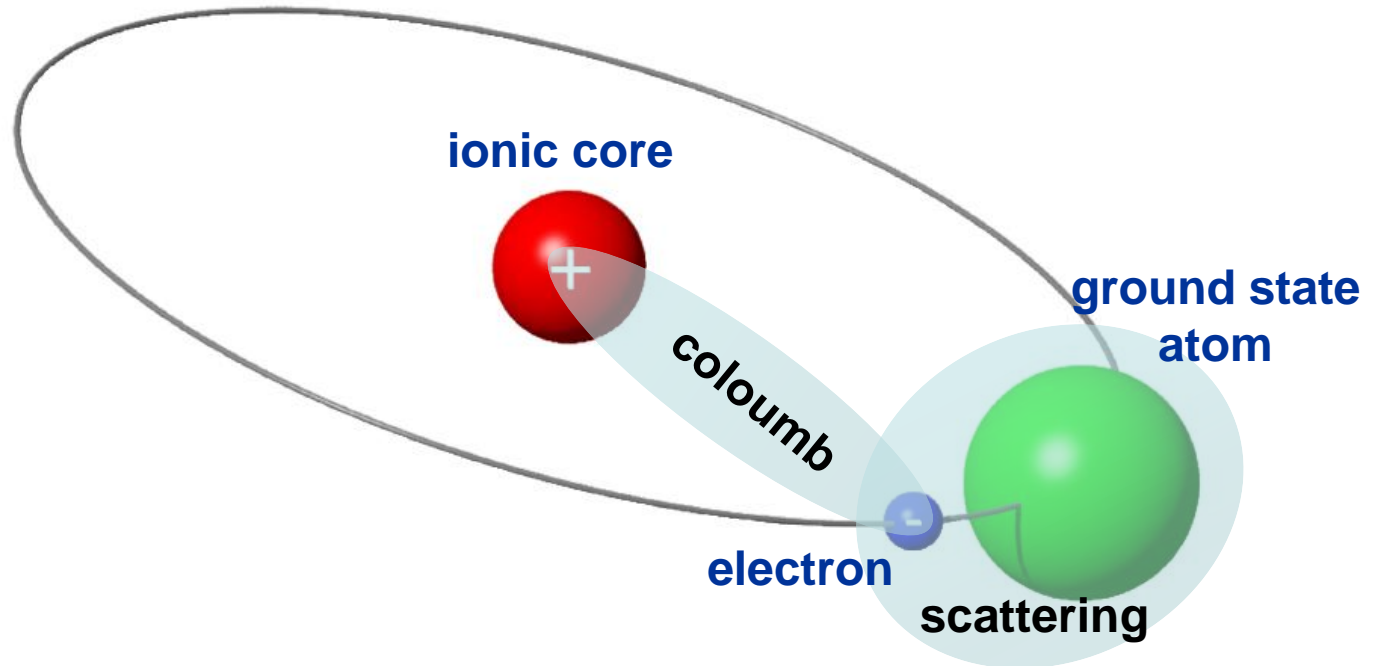


# Rydberg molecules



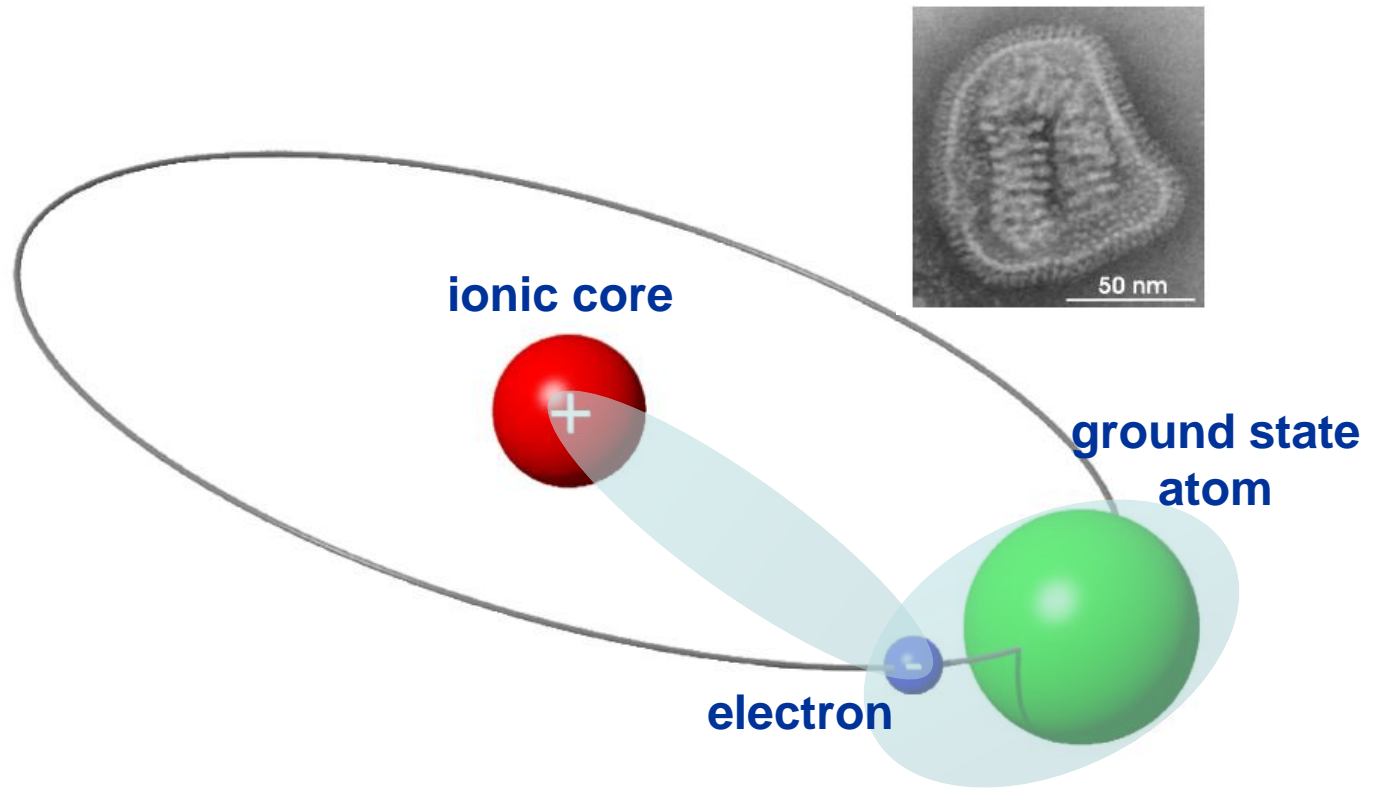


# Rydberg molecules





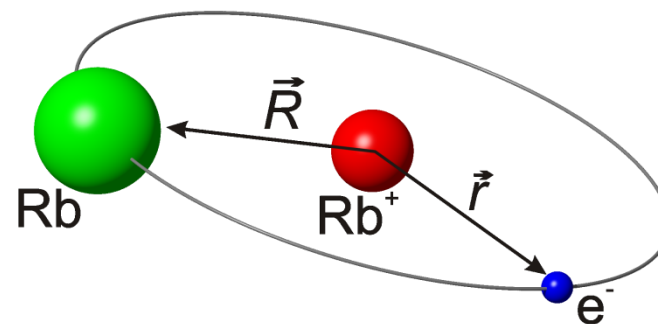
# Rydberg molecules



# Electron-atom scattering

neutral Rb polarized by electron

$$V_{pol}(\vec{r}, \vec{R}) = -\frac{1}{2}\alpha\vec{E}^2 \propto \frac{1}{|\vec{R} - \vec{r}|^4}$$



quantum mechanical s-wave scattering

- pseudopotential

$$V_{pseudo}(\vec{r}, \vec{R}) = 2\pi a(k)\delta(\vec{r} - \vec{R})$$

- scattering length

E. Fermi: Nuovo Cimento 11 (1934)

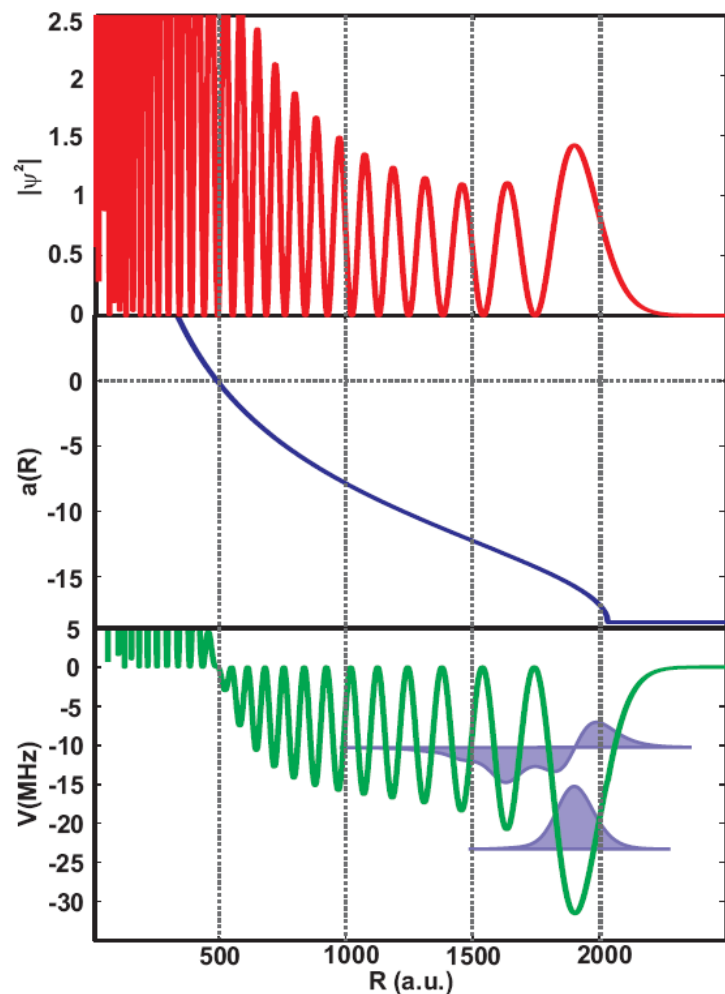
$$a[k(R)] = a_0 + \frac{\pi\alpha}{3}k(R) + O(k^2)$$

- classical k-dependence of Rydberg electron

$$\frac{1}{2}k^2(R) = -\frac{1}{2n^2} + \frac{1}{R}$$



# Simplest possible model for the molecular potential



probability density

$$|\psi_{35,0,0}|^2(\mathbf{R})$$



energy-dependent scattering length

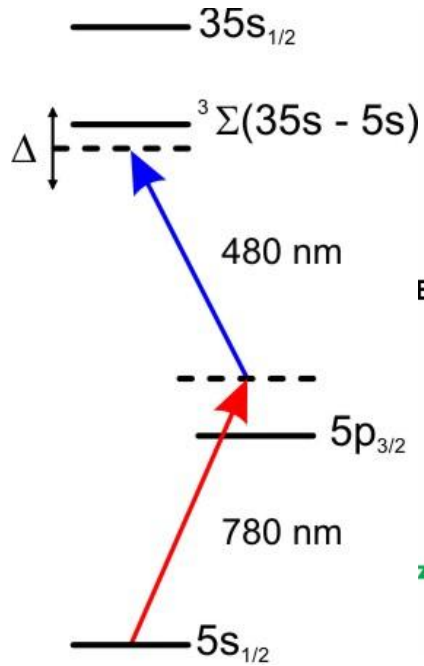
$$a[k(R)] = a_0 + \frac{\pi\alpha}{3}k(R)$$



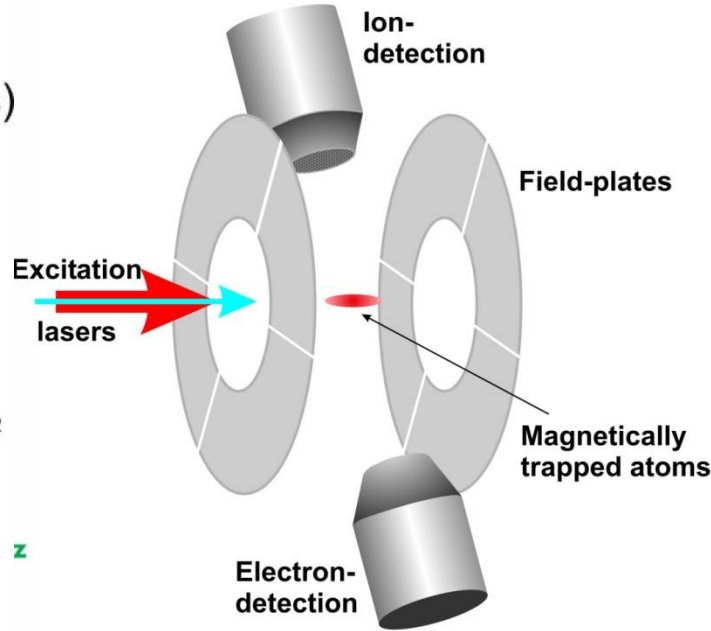
molecular potential

$$V_0(\mathbf{R}) = 2\pi a[k(\mathbf{R})] |\Psi_{35,0,0}(\mathbf{R})|^2$$

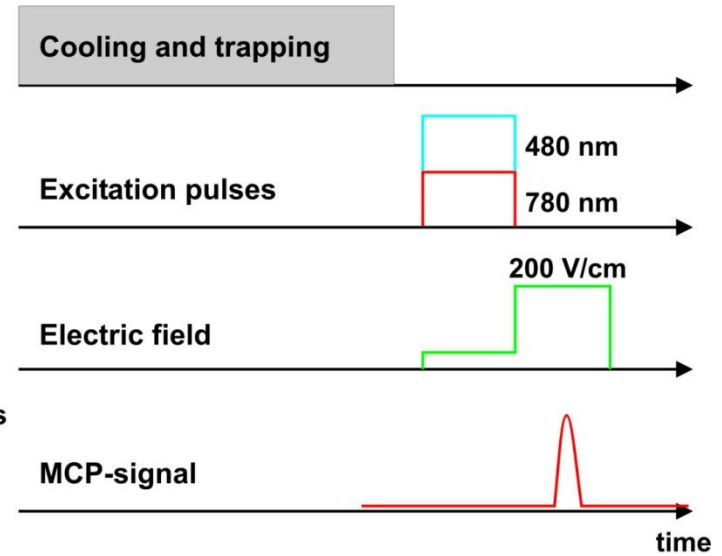
# Some experimental details



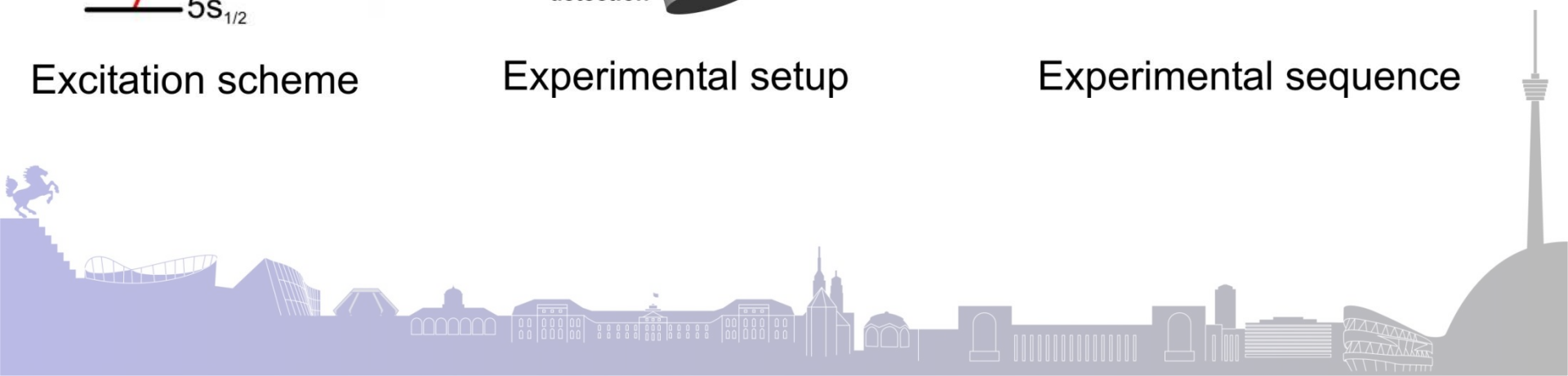
Excitation scheme



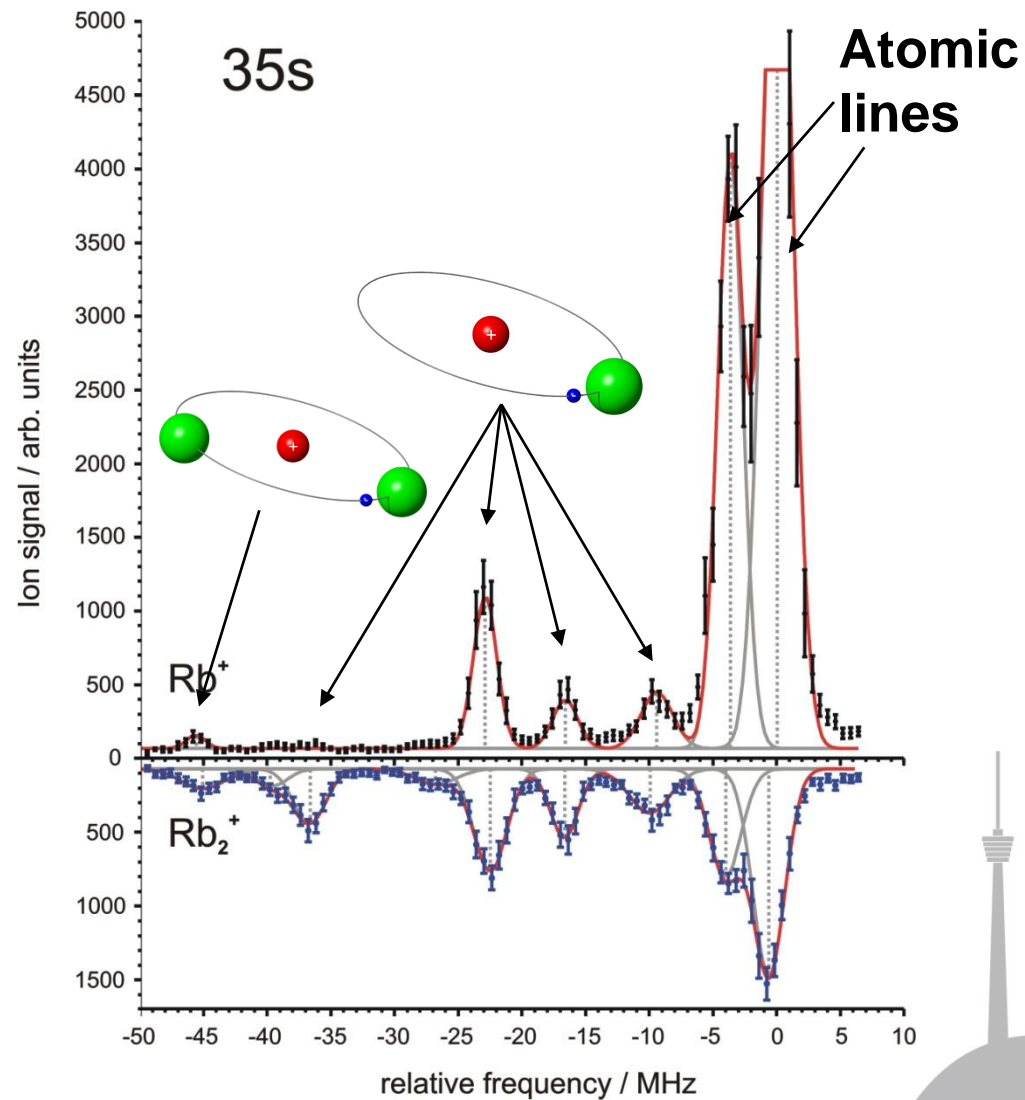
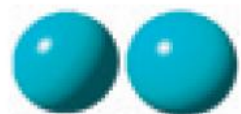
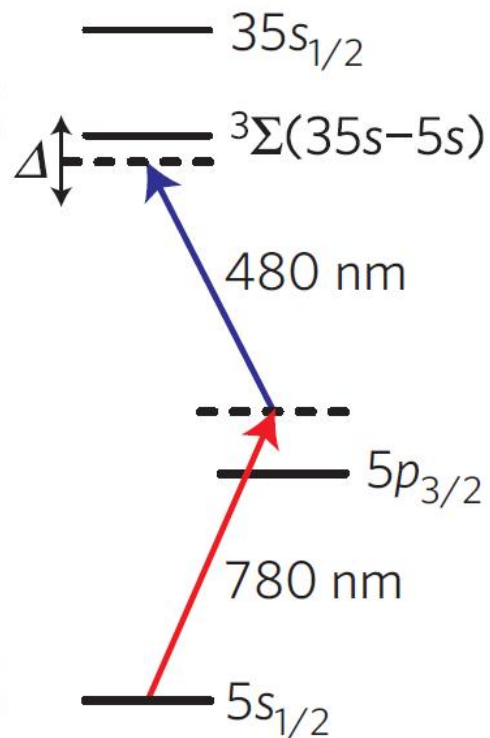
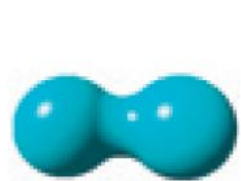
Experimental setup



Experimental sequence

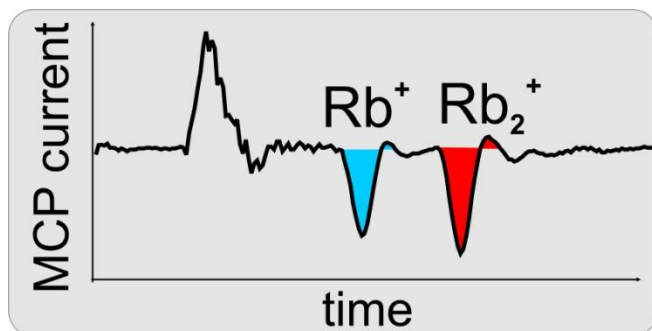


# Photo association of Rydberg molecules



# Two kinds of ions signals

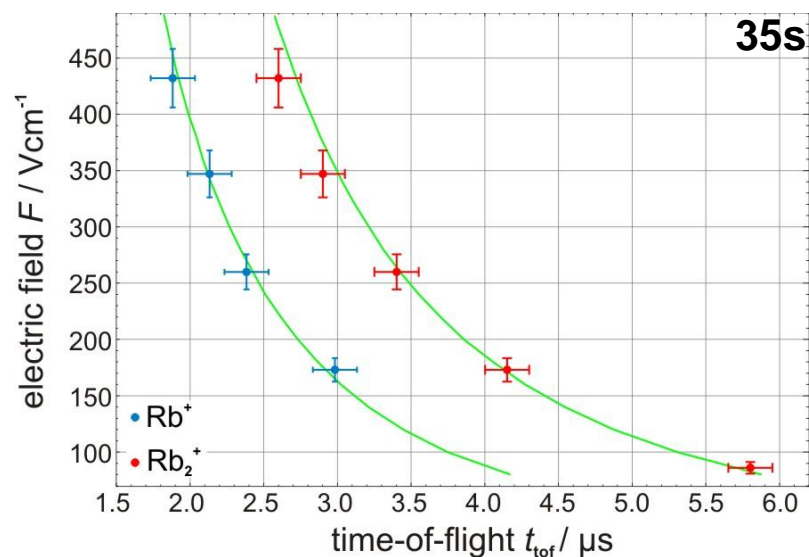
detected ion signal



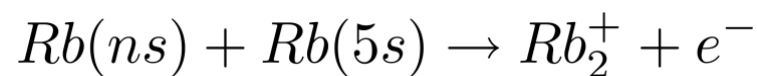
two signals:

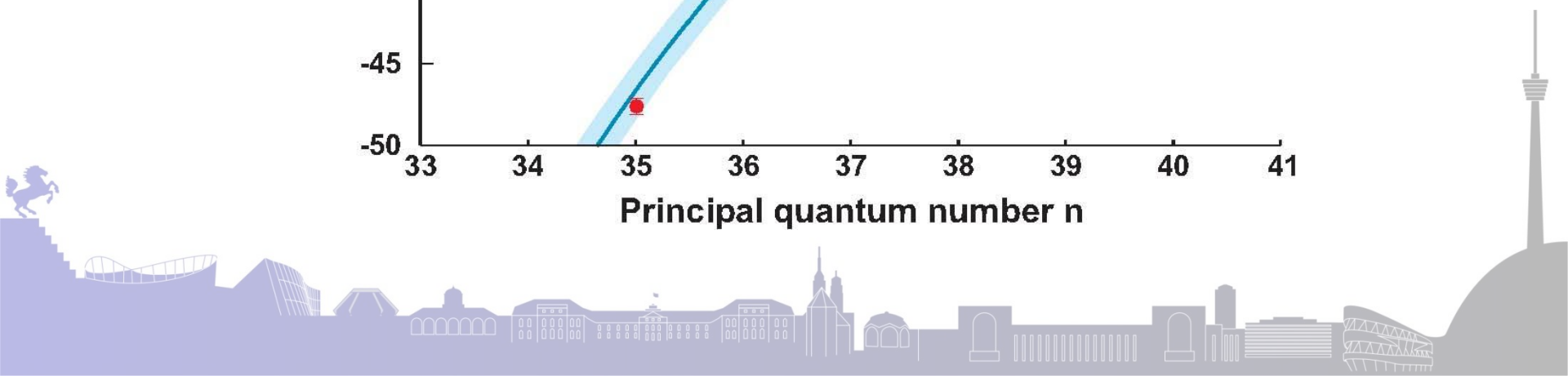
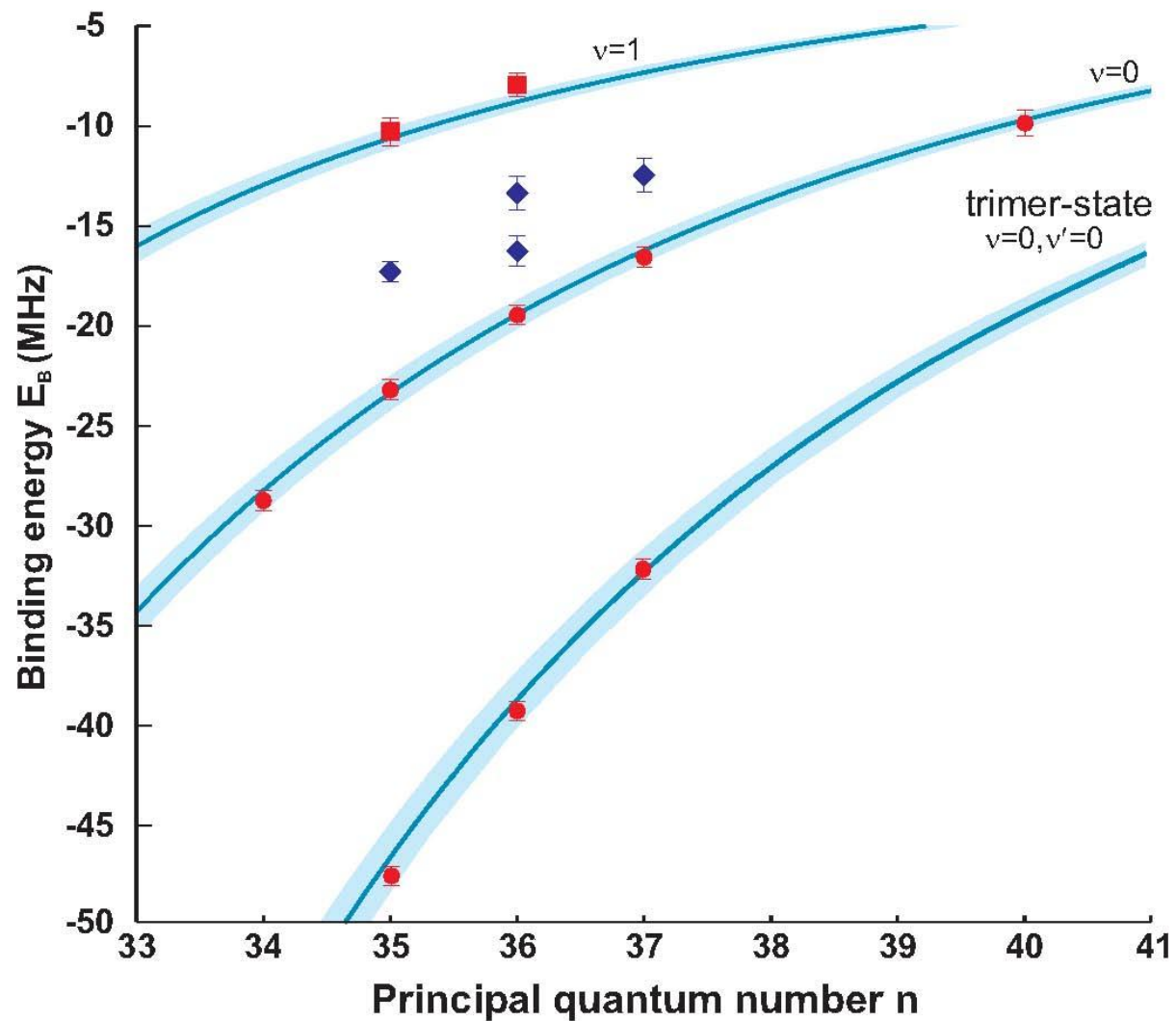
- atomic ion signal  $Rb^+$
- molecular ion signal  $Rb_2^+$

time-of-flight measurement



source of  $Rb_2^+$ :



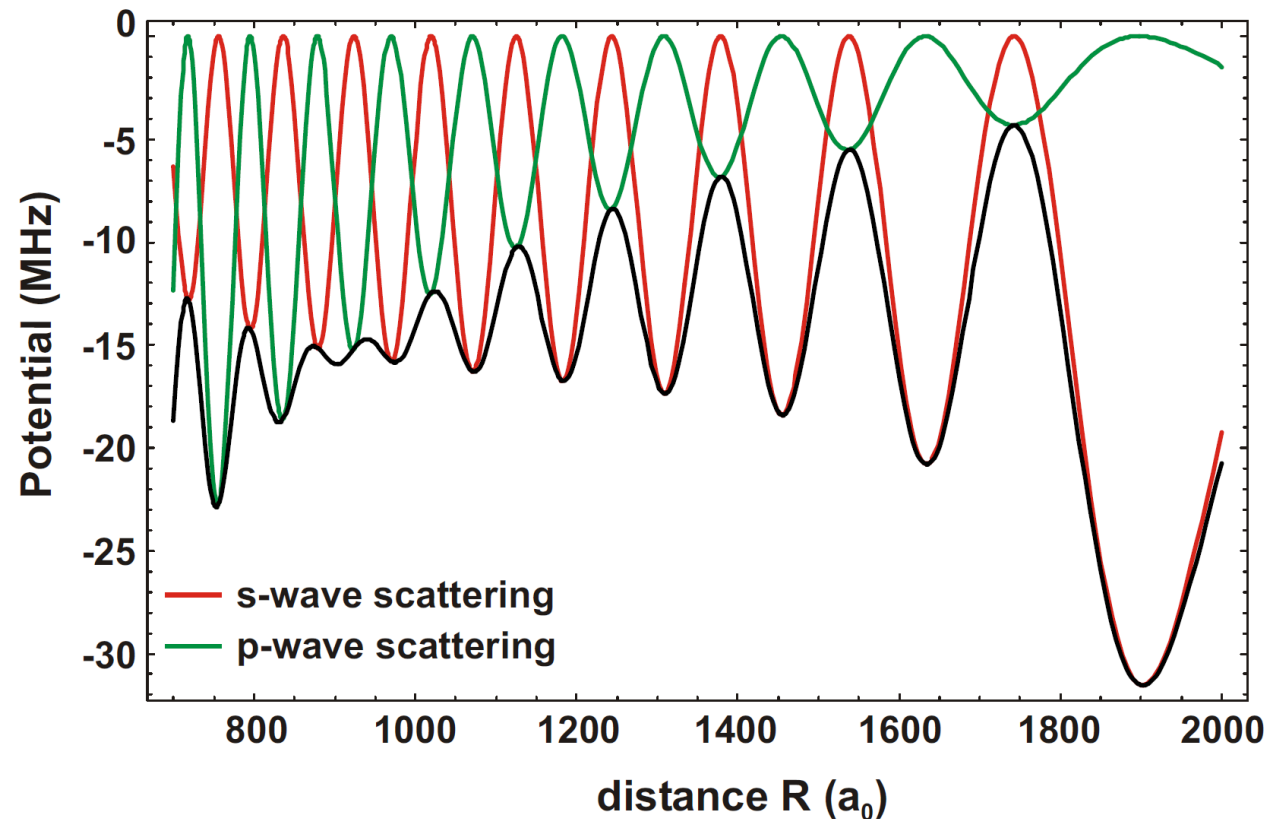




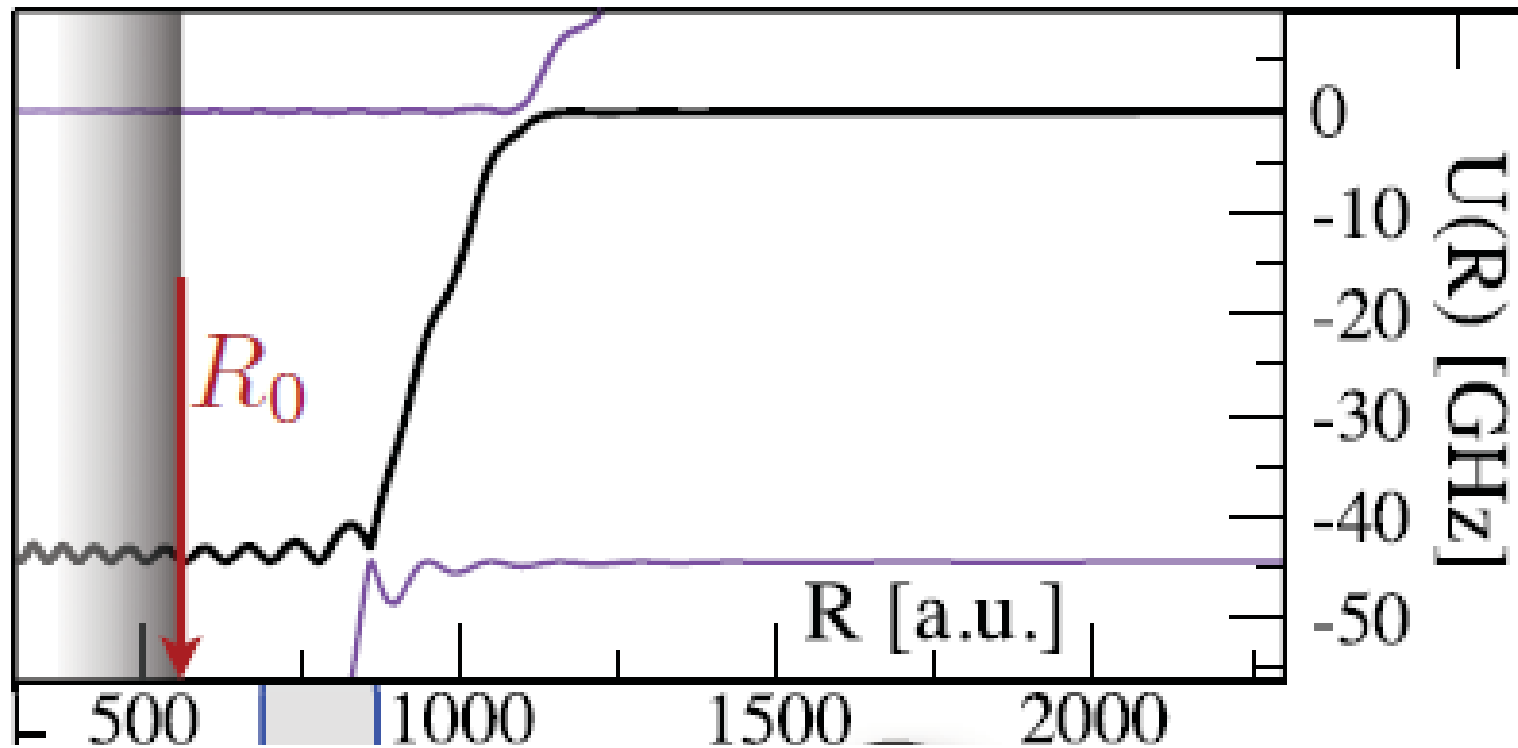
## A closer look on the potential

s-wave potential  $V_0(\vec{\mathbf{R}}) = 2 \pi a [\mathbf{k}(\mathbf{R})] \cdot \left| \Psi_{n,l,m}(\vec{\mathbf{R}}) \right|^2$

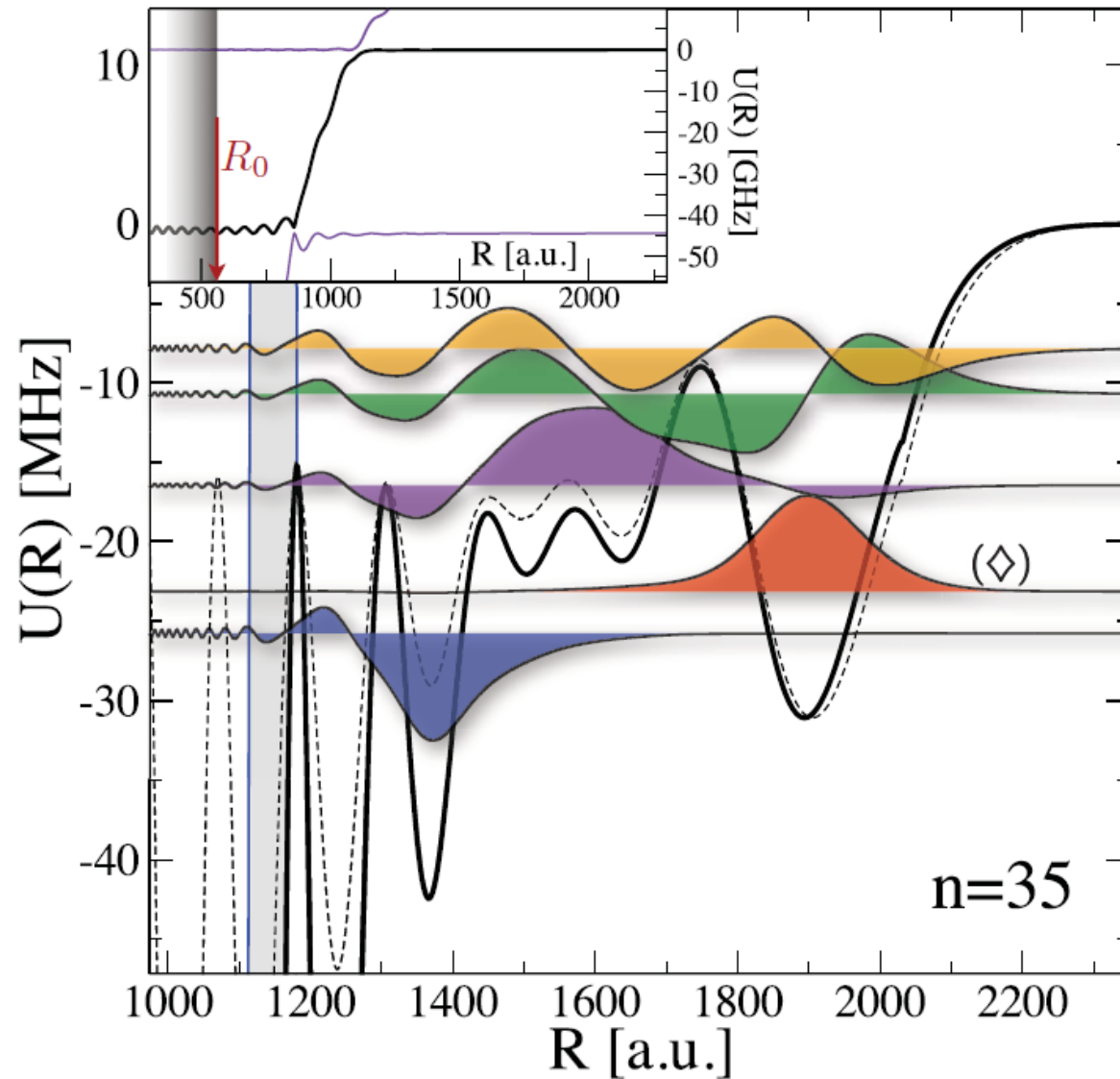
p-wave potential  $V_1(\vec{\mathbf{R}}) = -\frac{2 \pi^2 \alpha}{5 \mathbf{k}(\mathbf{R})} \cdot \left| \nabla \Psi_{n,l,m}(\vec{\mathbf{R}}) \right|^2$  Omont, J. Physique **38**, 1343 (1977)



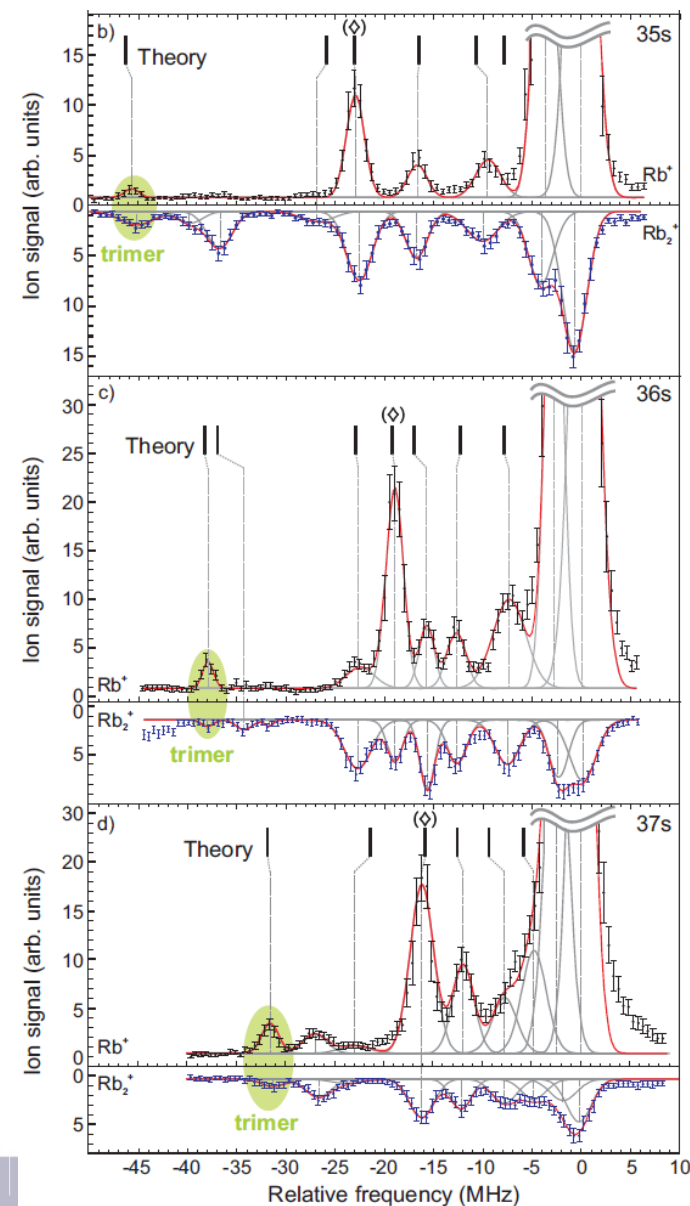
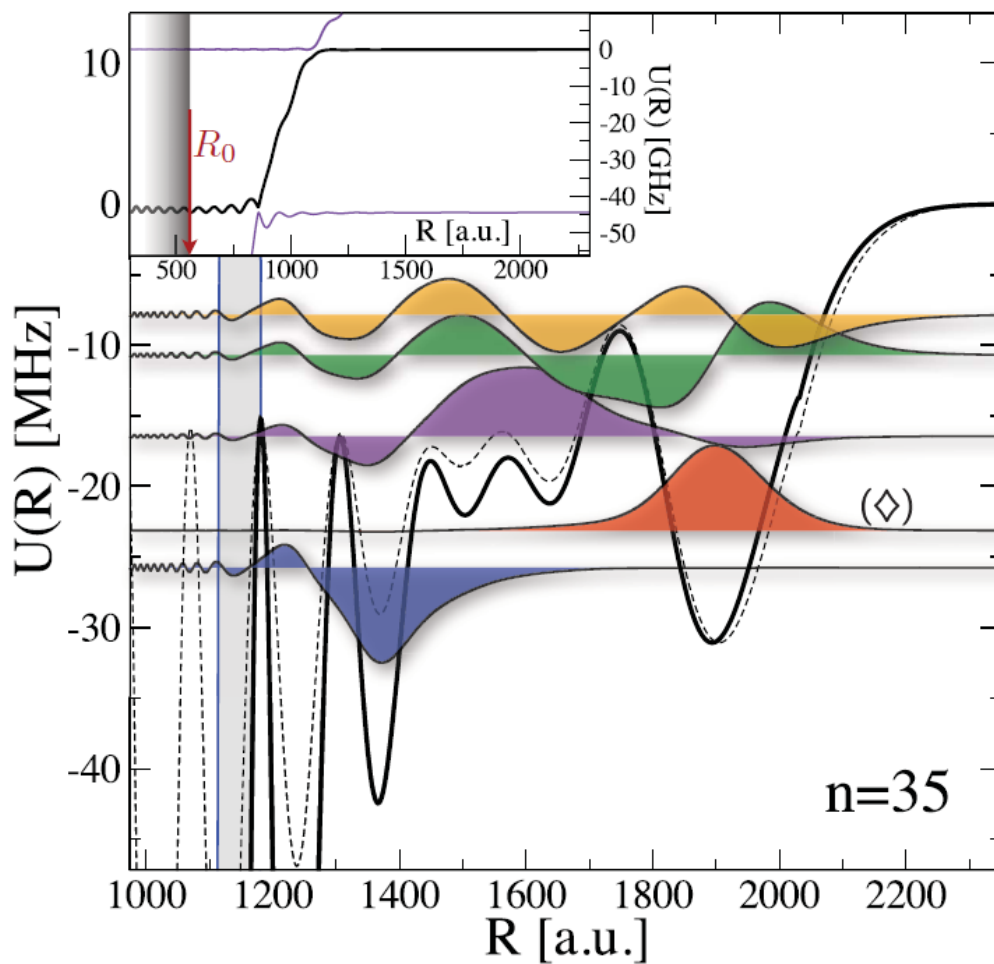
## A wider look on the potential



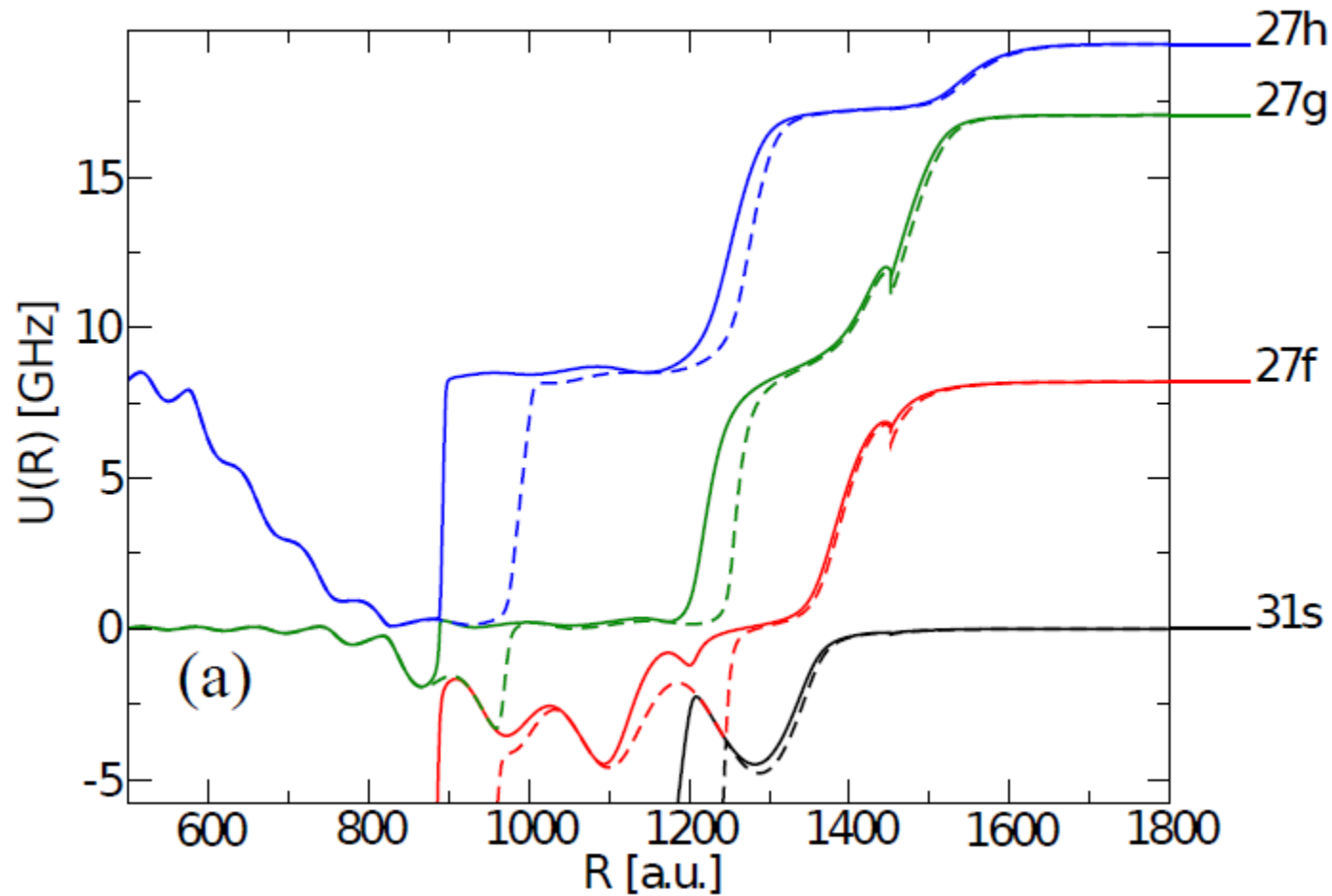
# Exact calculation of the potential



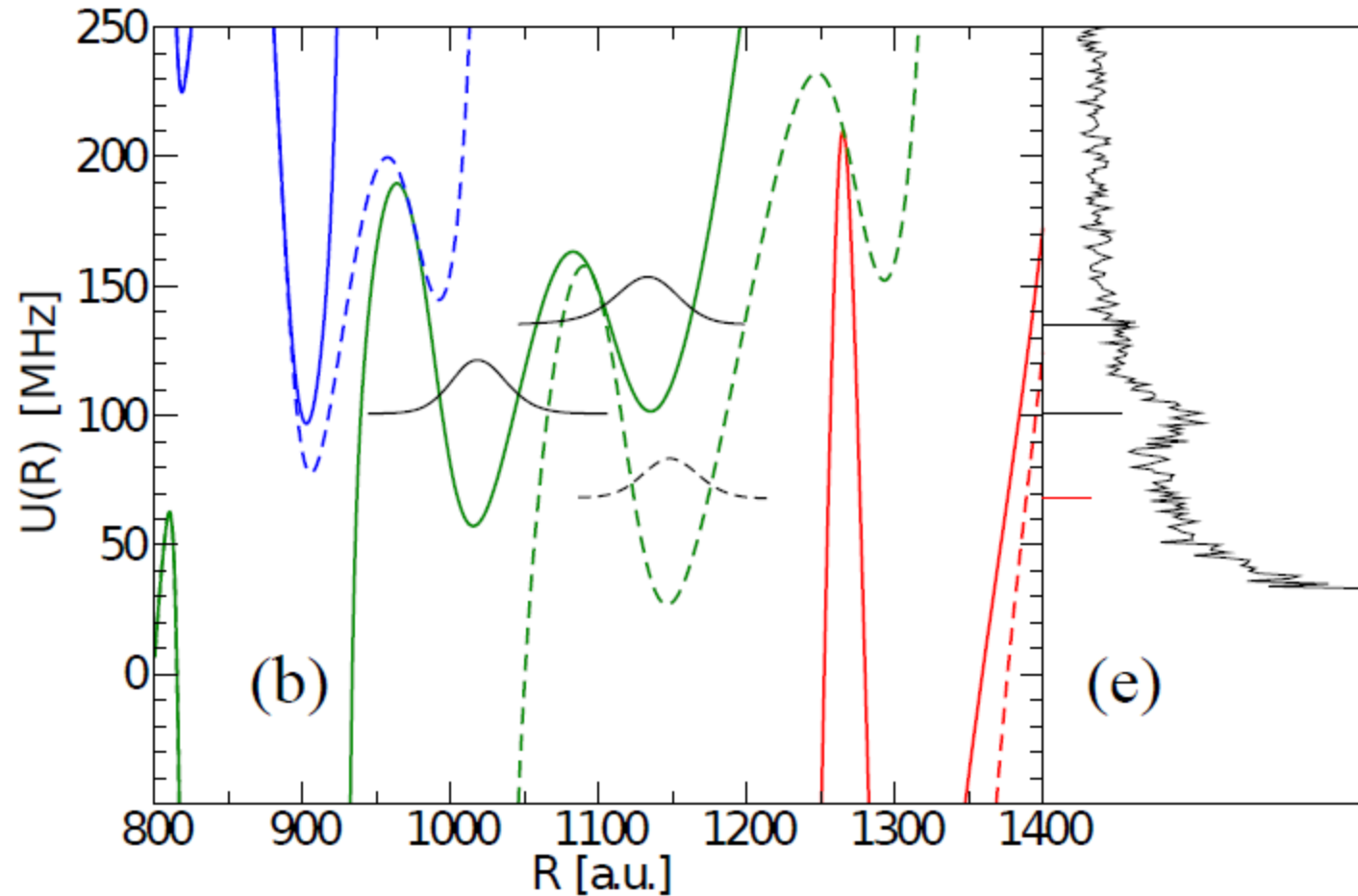
# Exact calculation of the potential



# How about Cesium

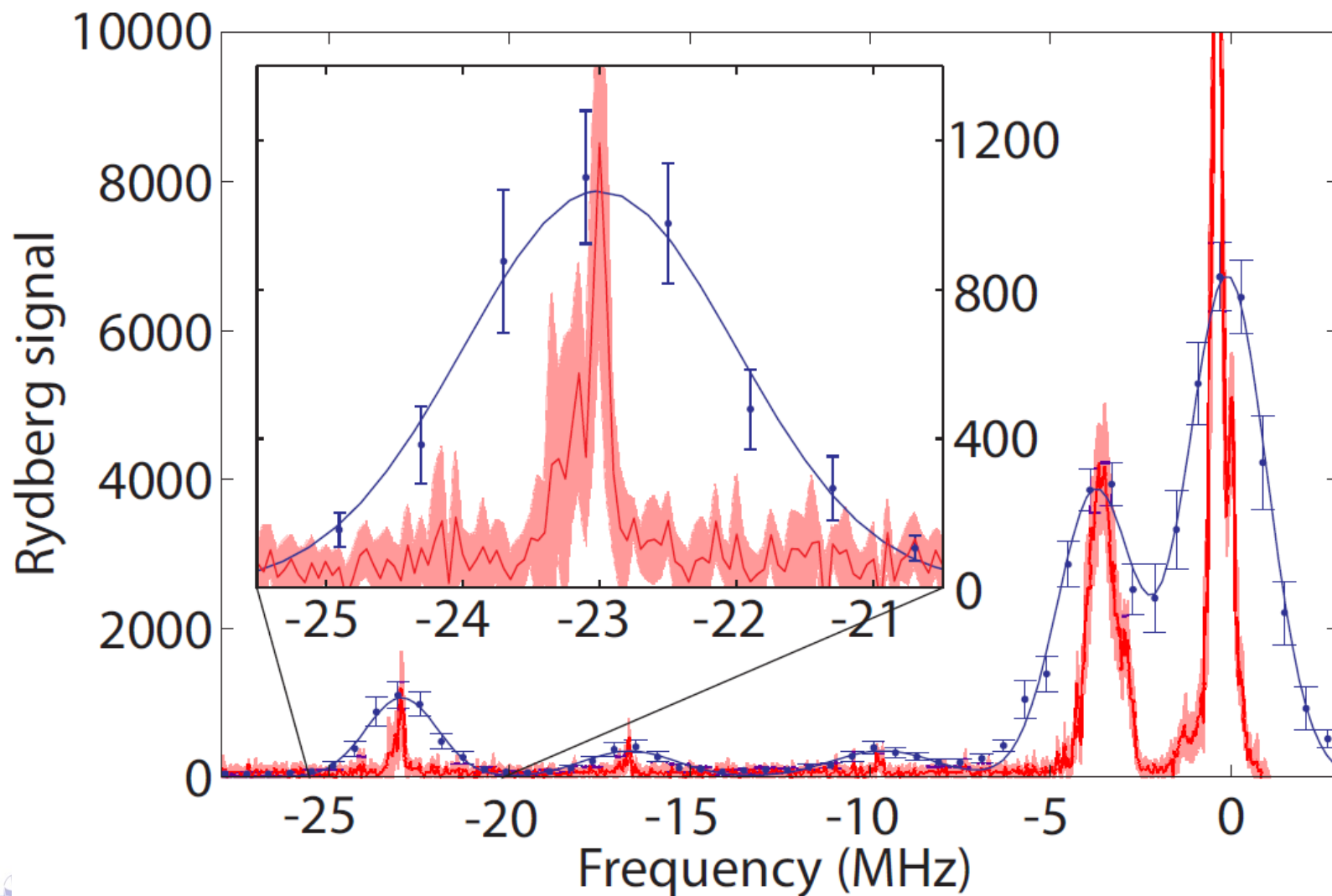


# How about Cesium



Shaffer, Arxiv1205.4974

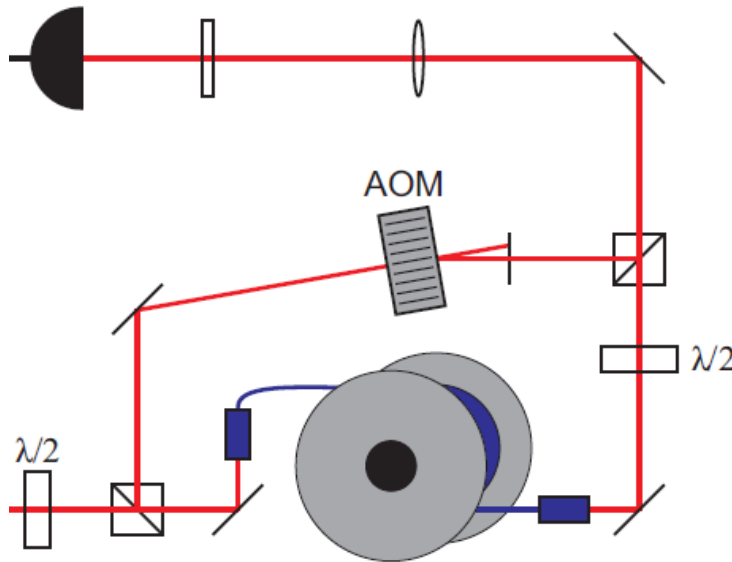
# High resolution photoassociation spectrum



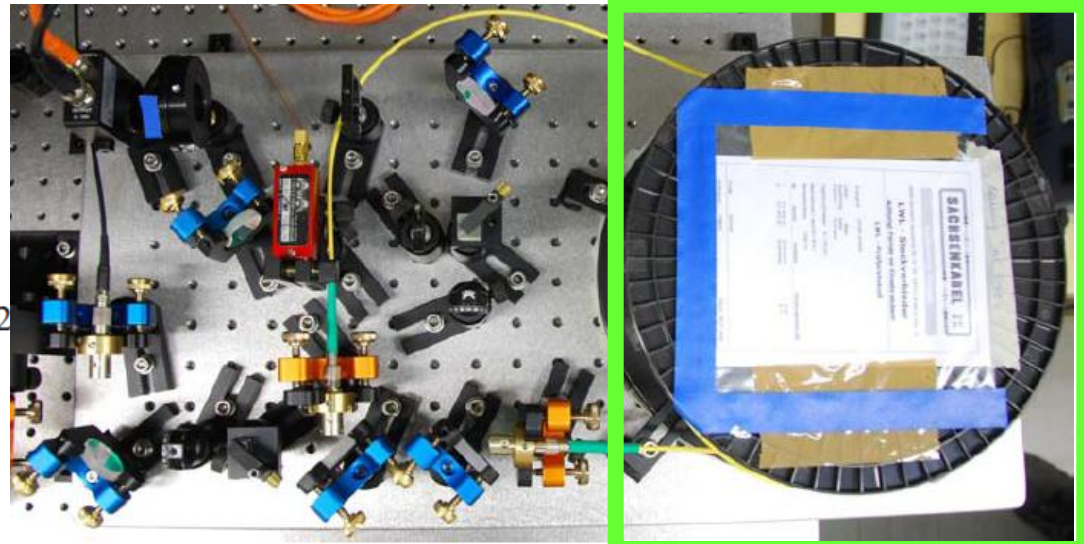


# Characterization by selfheterodyne detection

Fast PD



10 km fiber  
 $\tau=52\mu\text{s}$

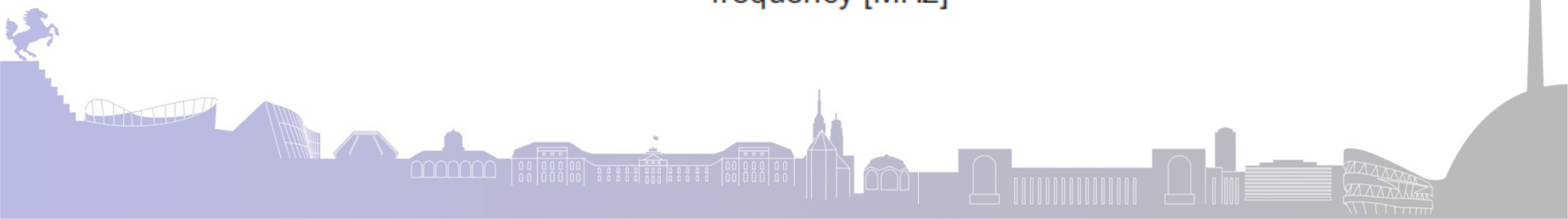
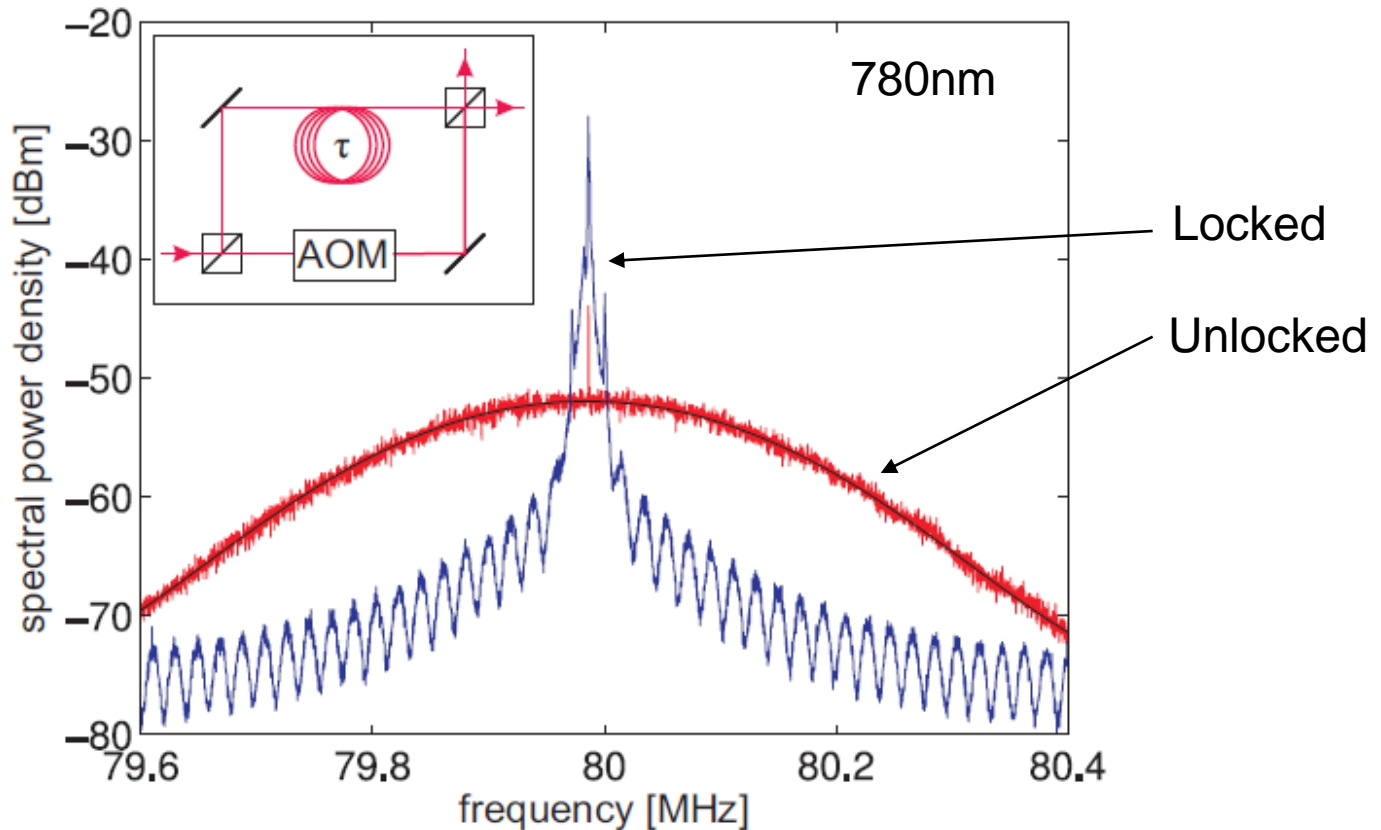


Acoustic shielding  
(foam) necessary!

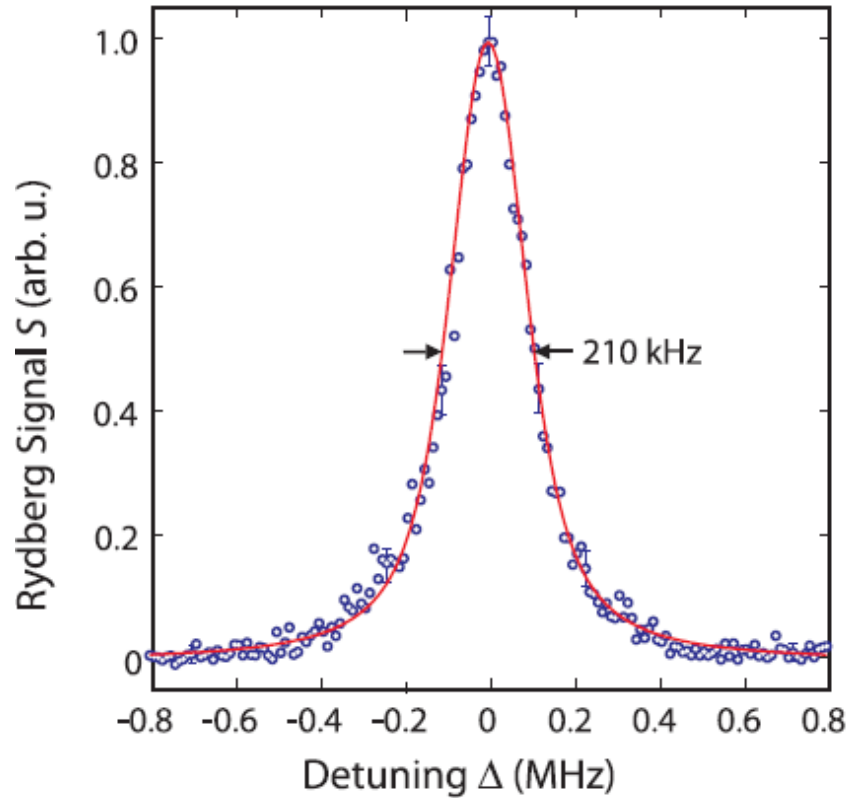




# Characterization by selfheterodyne detection

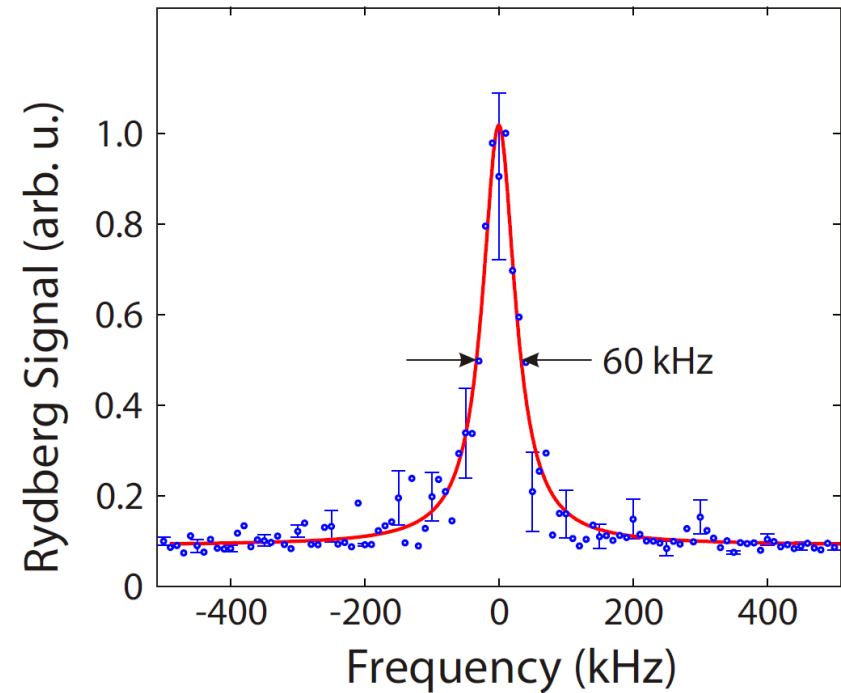


## Rydberg molecules



**Voigt Fit for fixed 3  $\mu\text{K}$ :  
142 kHz Lorentz linewidth**

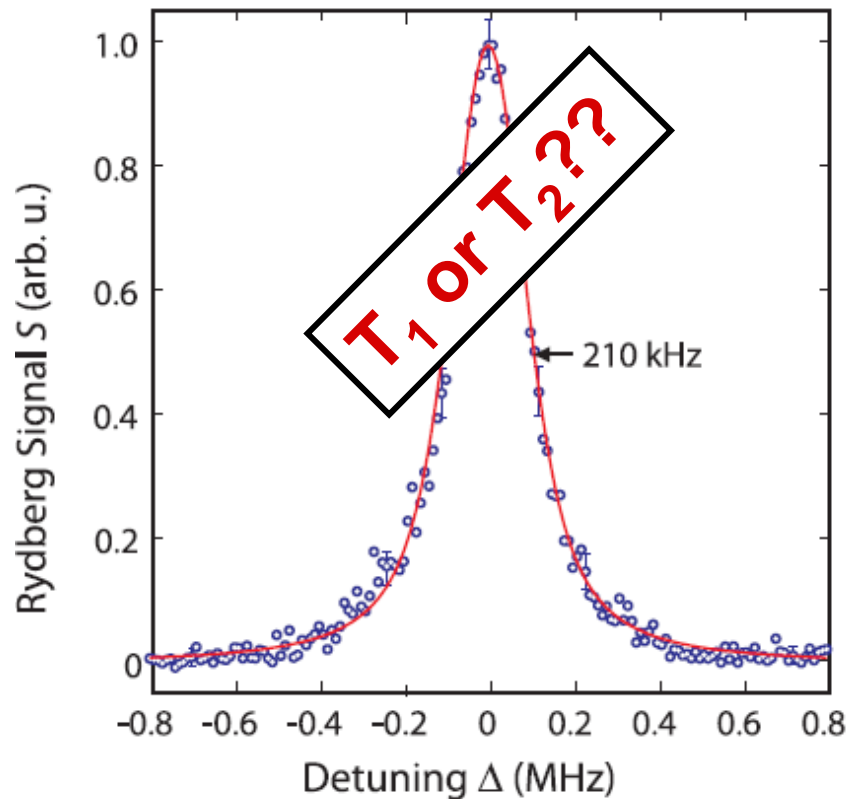
## Rydberg atoms



**Limited by Laser linewidth**

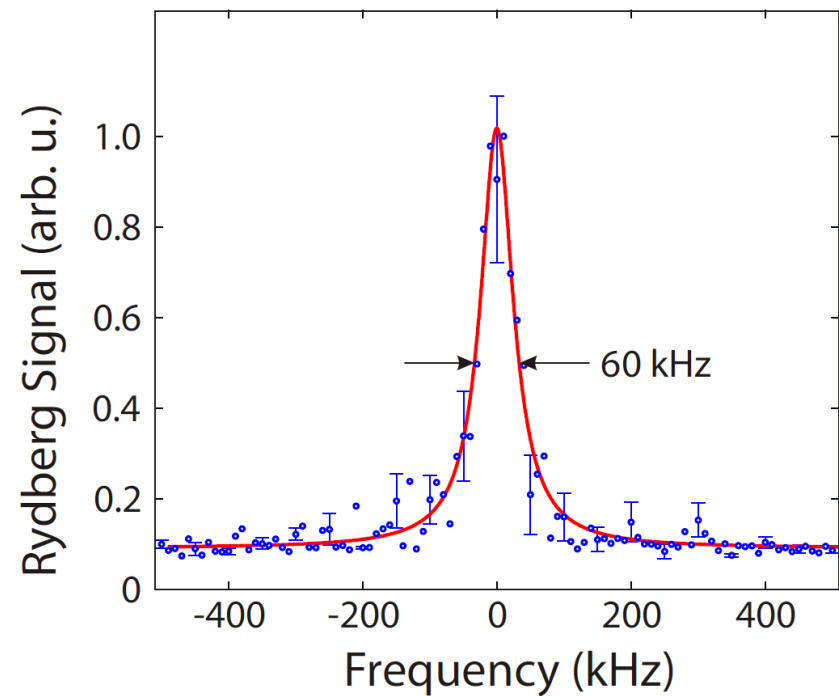


## Rydberg molecules

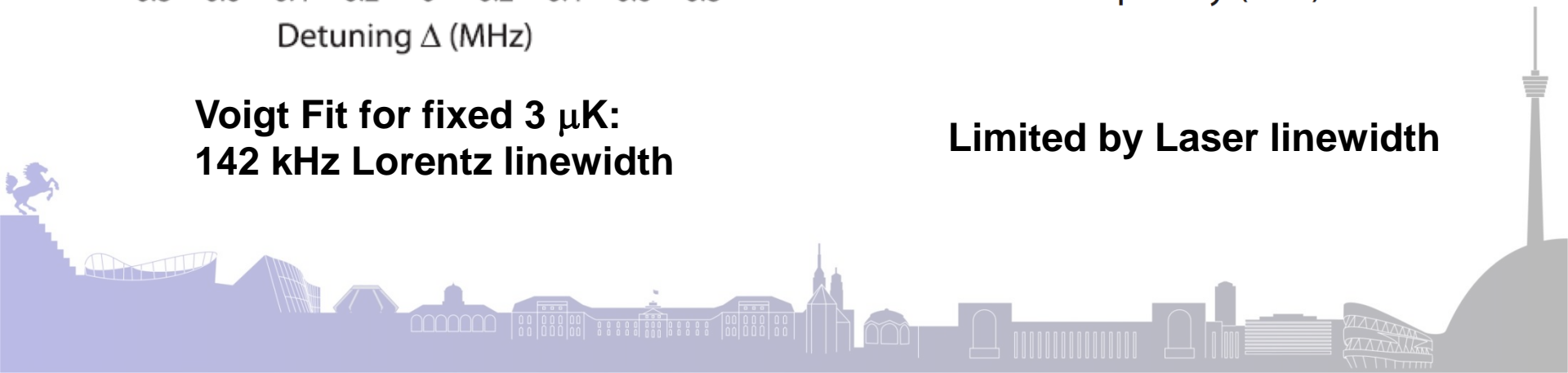


**Voigt Fit for fixed 3  $\mu$ K:  
142 kHz Lorentz linewidth**

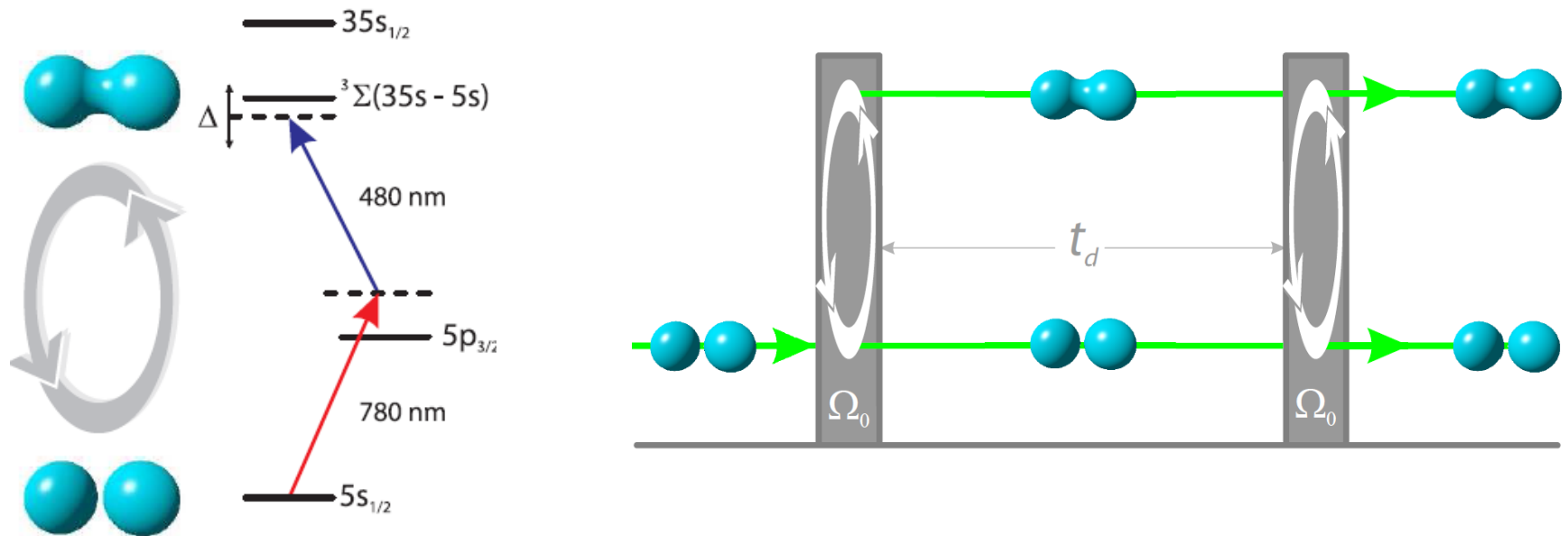
## Rydberg atoms



**Limited by Laser linewidth**

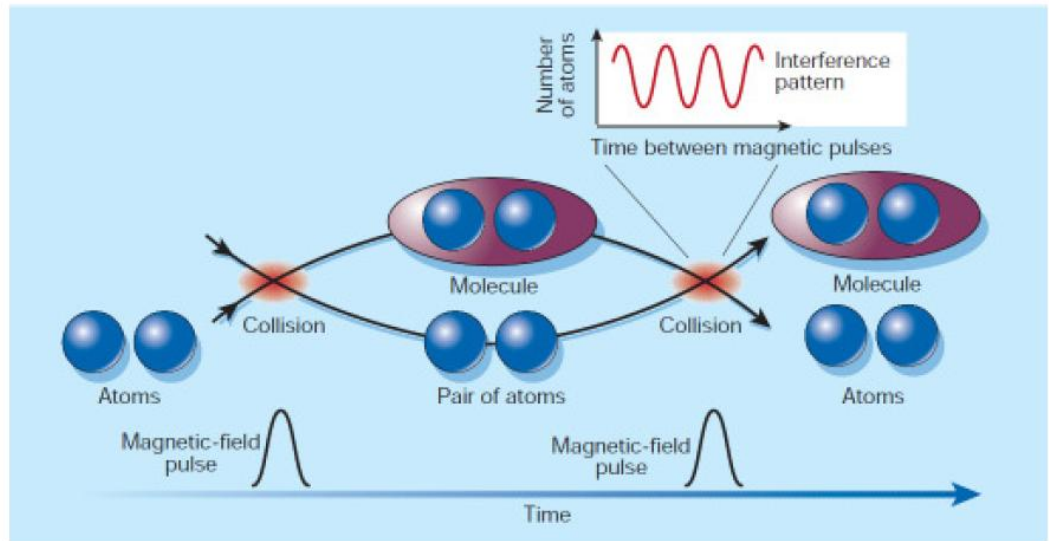
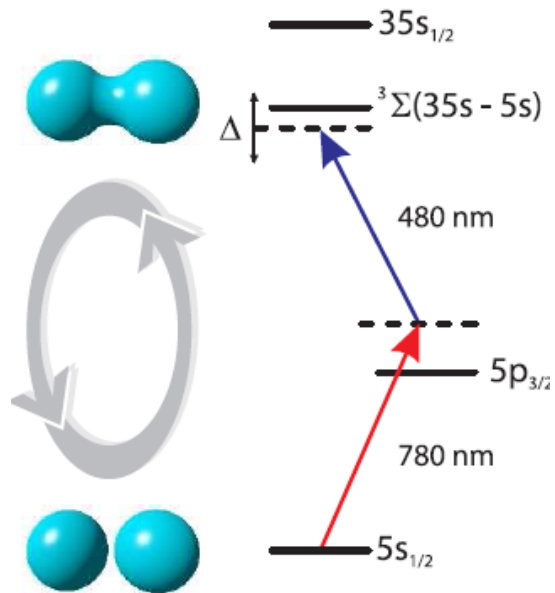


# Ramsey interferometry with Rydberg molecules





# Ramsey interferometry with Rydberg molecules



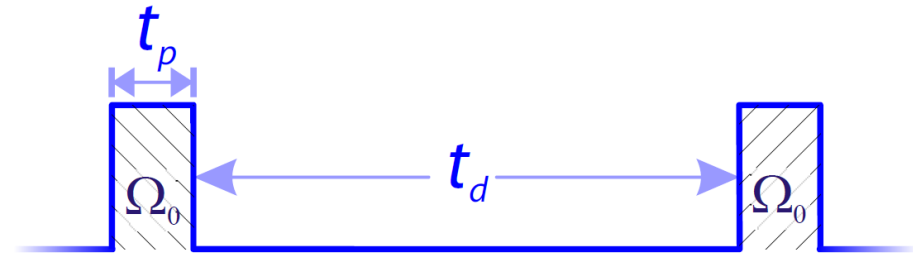
P. Zoller, Nature, **417** 493 (2002)

First experiment with Feshbach molecules (JILA) Nature, **417** 529 (2002)



# Ramsey fringes: (cf. Young's double slit)

separate two pulses by  
variable delay time:

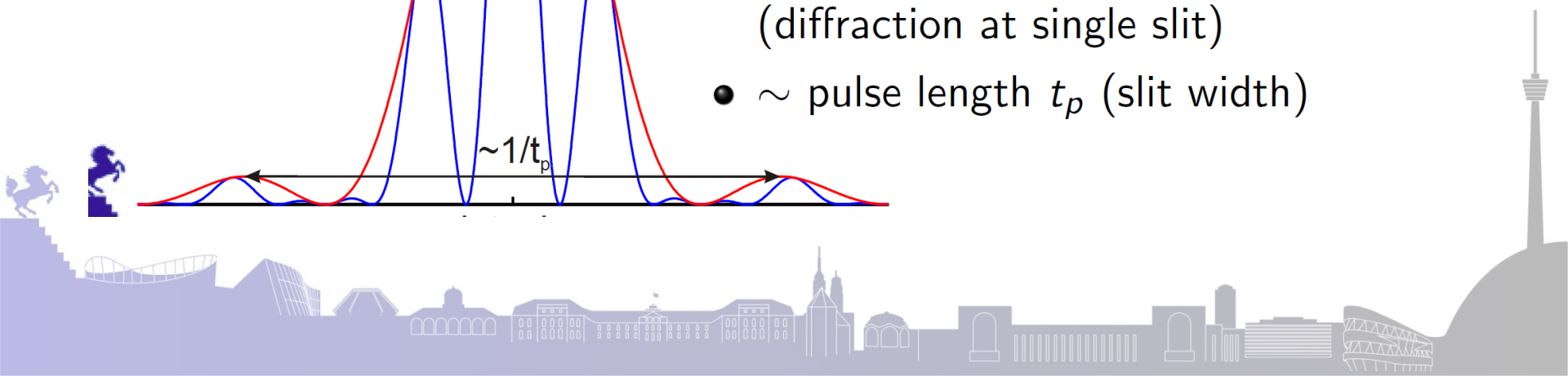
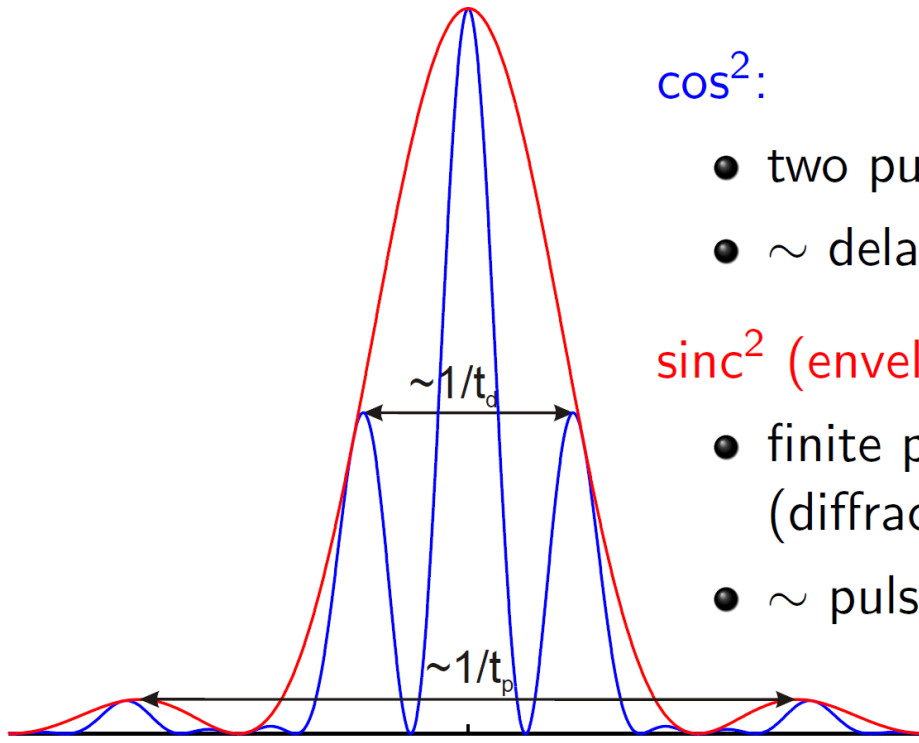


$\cos^2$ :

- two pulses (interference of two slits)
- $\sim$  delay time  $t_d$  (slit distance)

$\text{sinc}^2$  (envelope):

- finite pulse length  
(diffraction at single slit)
- $\sim$  pulse length  $t_p$  (slit width)

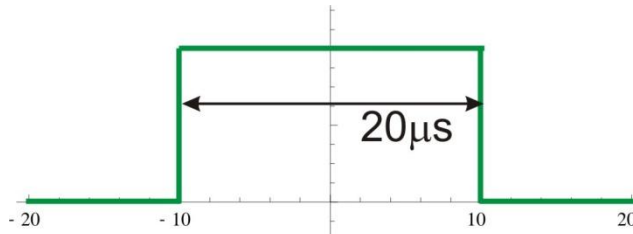




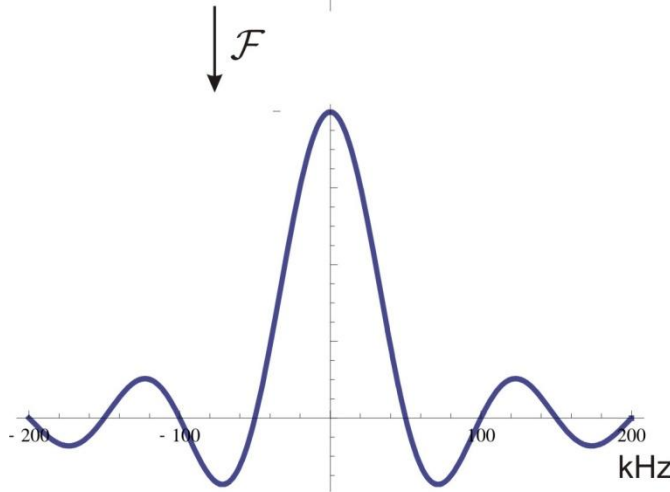


# Excitation time broadening

Pulse shape

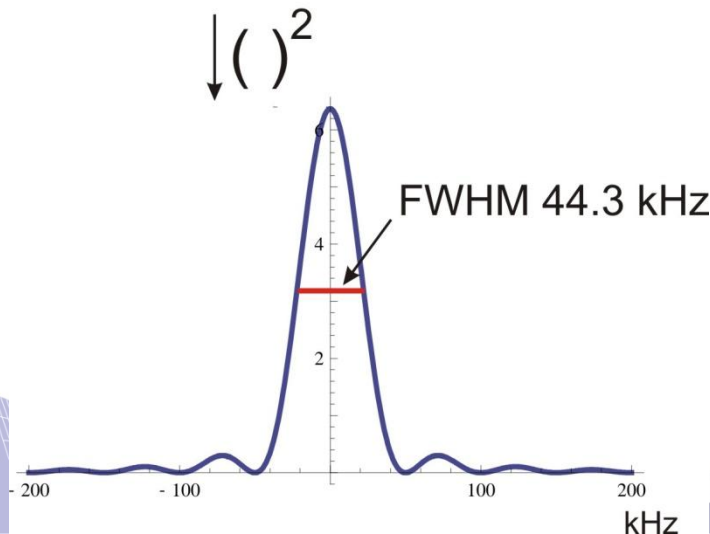


Amplitude of electric field

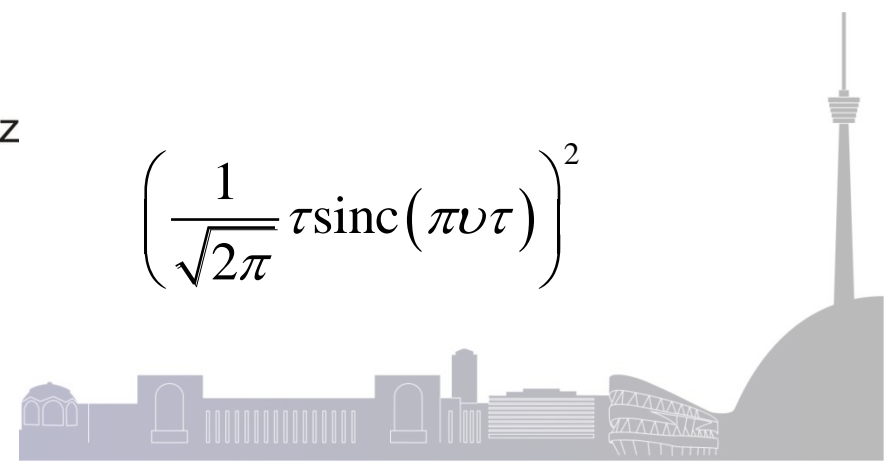


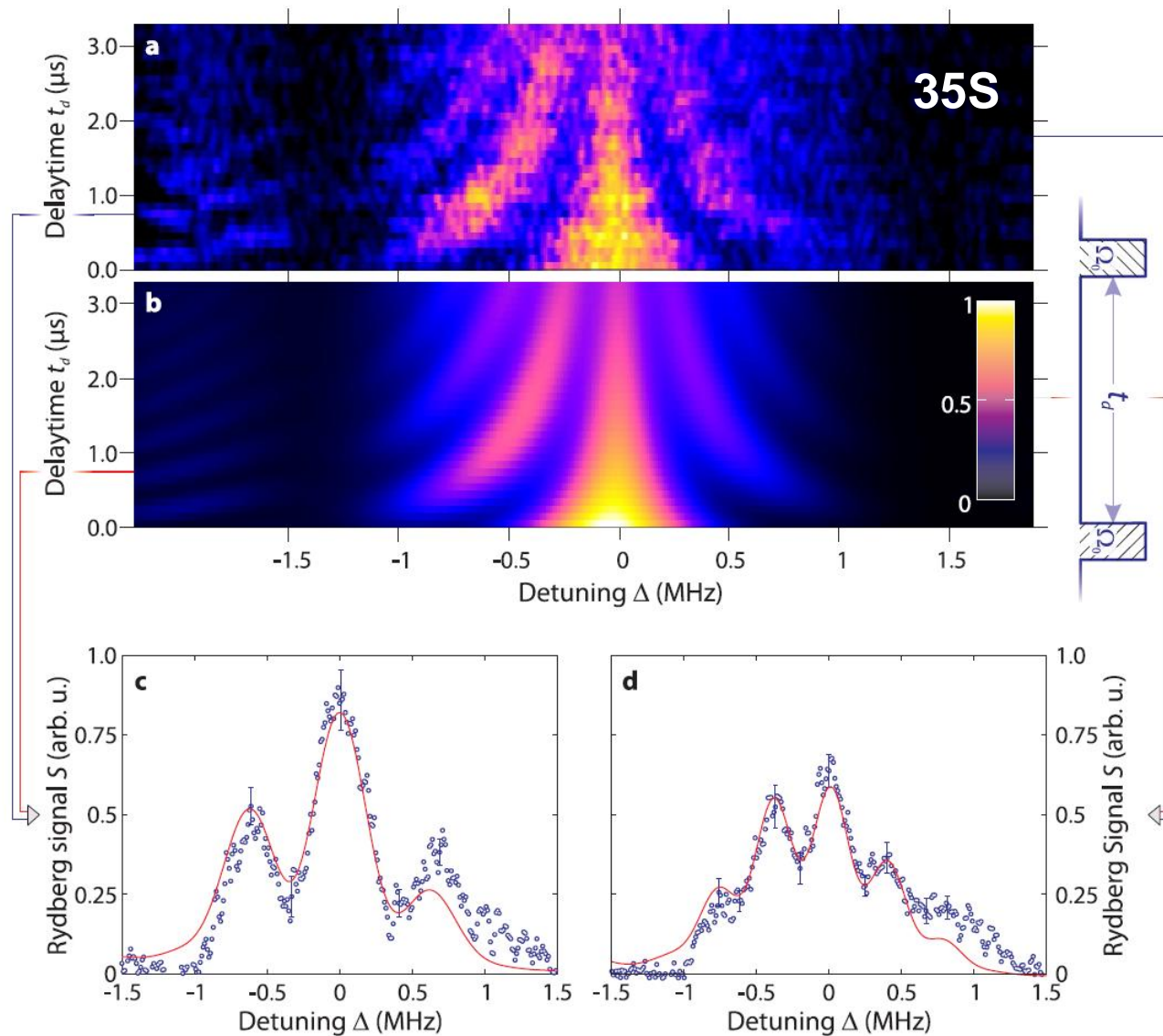
$$\frac{1}{\sqrt{2\pi}} \tau \text{sinc}(\pi \nu \tau)$$

Intensity of light field



$$\left( \frac{1}{\sqrt{2\pi}} \tau \text{sinc}(\pi \nu \tau) \right)^2$$

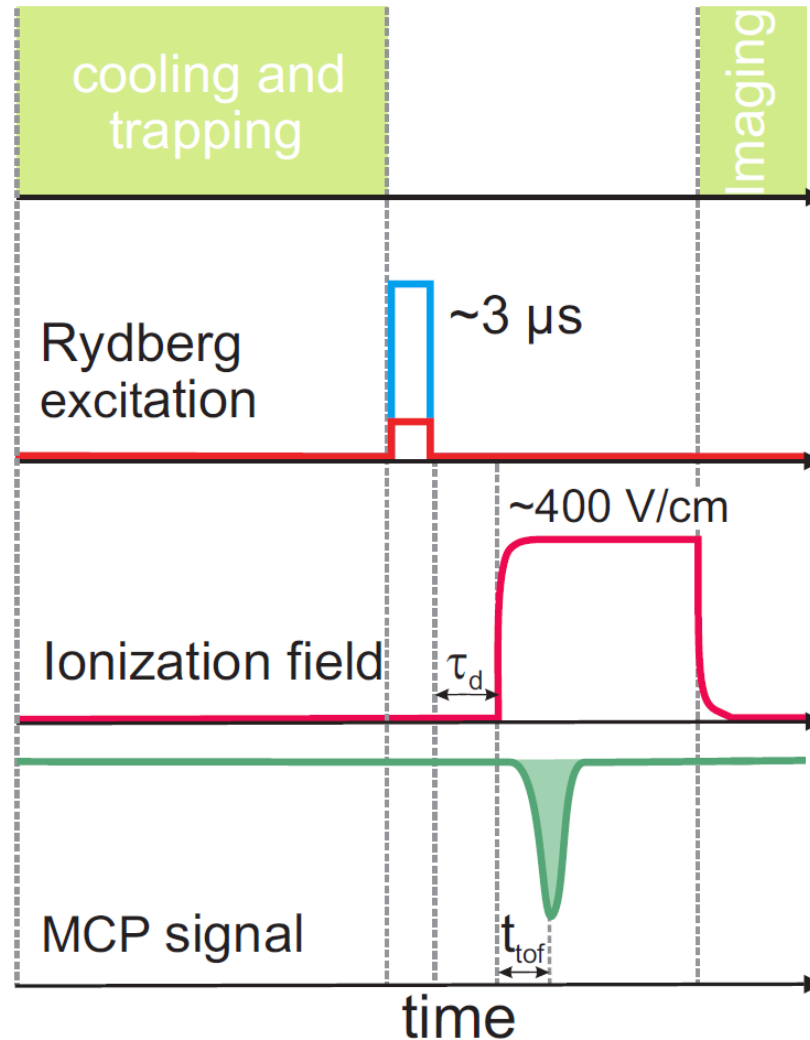




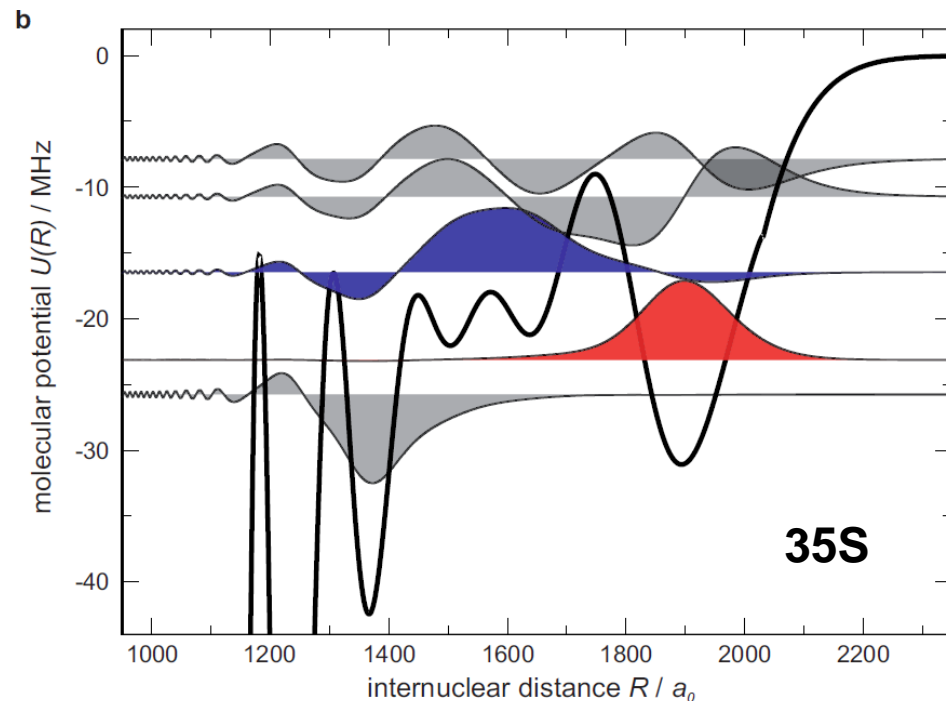
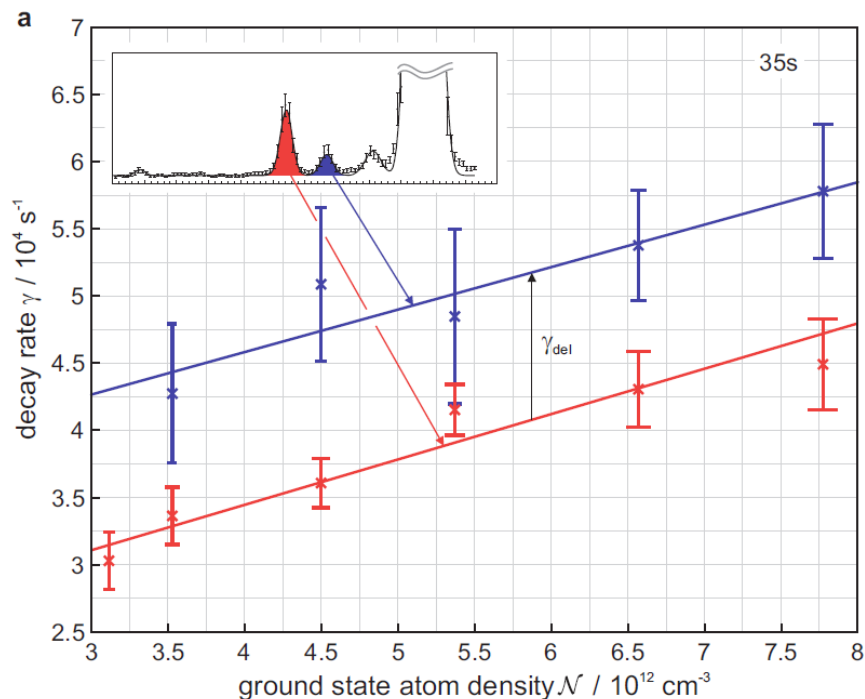
$$T_1 = 6.4(9) \mu\text{s}$$

$$T_2 = 1.5(2) \mu\text{s}$$

# Lifetimes of Rydberg molecules



# Lifetimes of Rydberg molecules

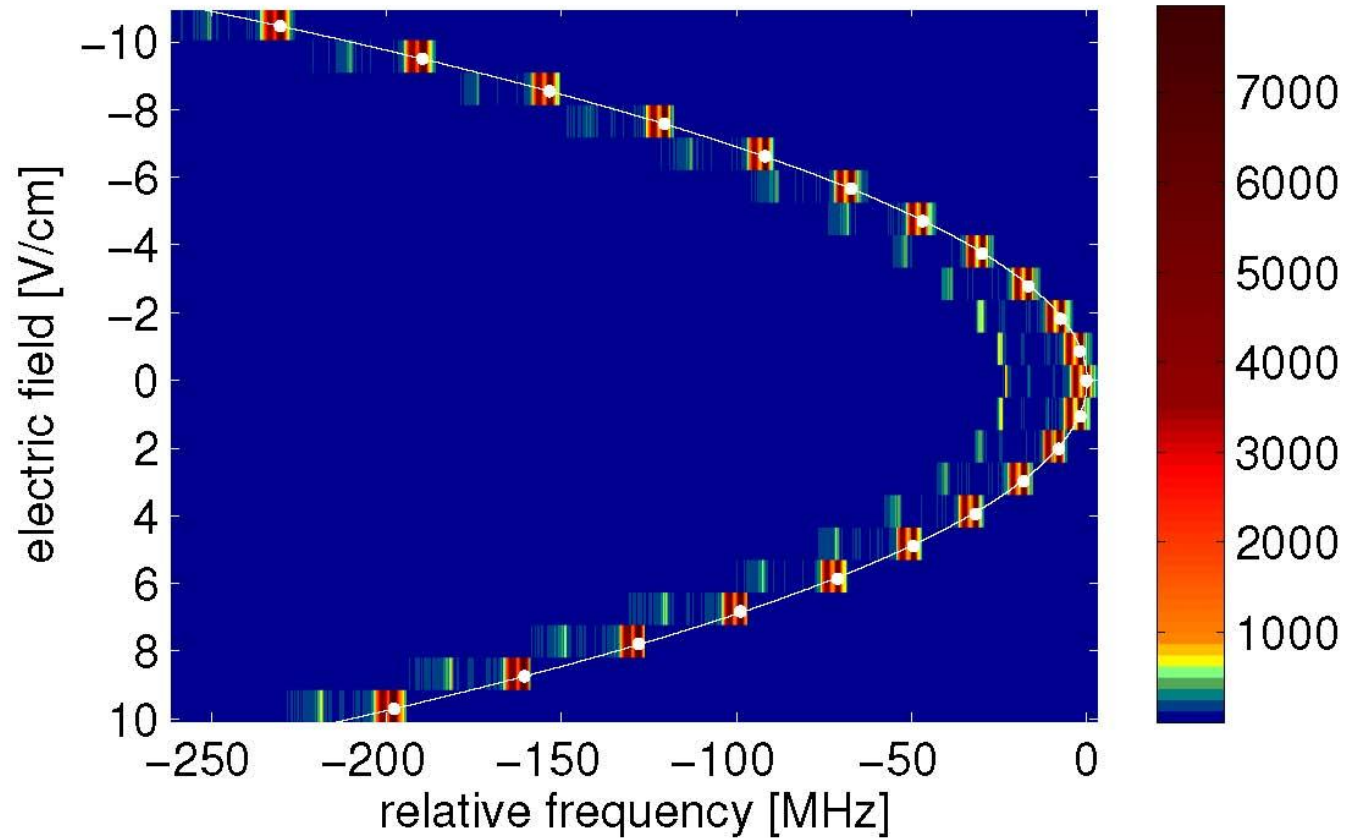


$$\gamma = \gamma_{\text{radiative}} + \underbrace{\gamma_{\text{collision}}}_{\text{NOV}} + \gamma_{\text{transmission}}$$

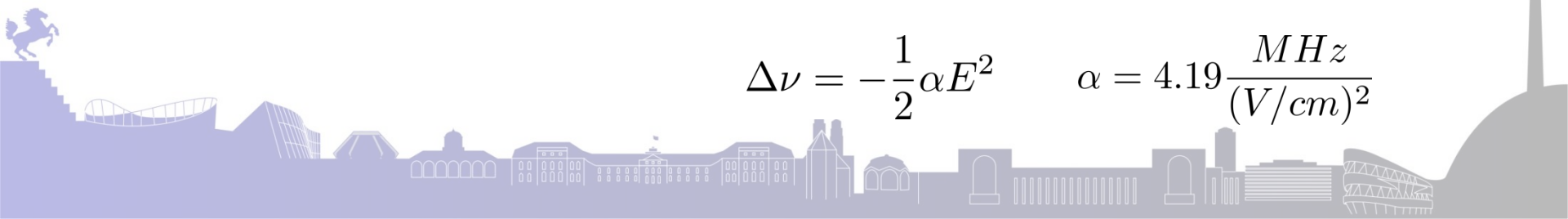
J. Phys. B. 44 184004 (2011)



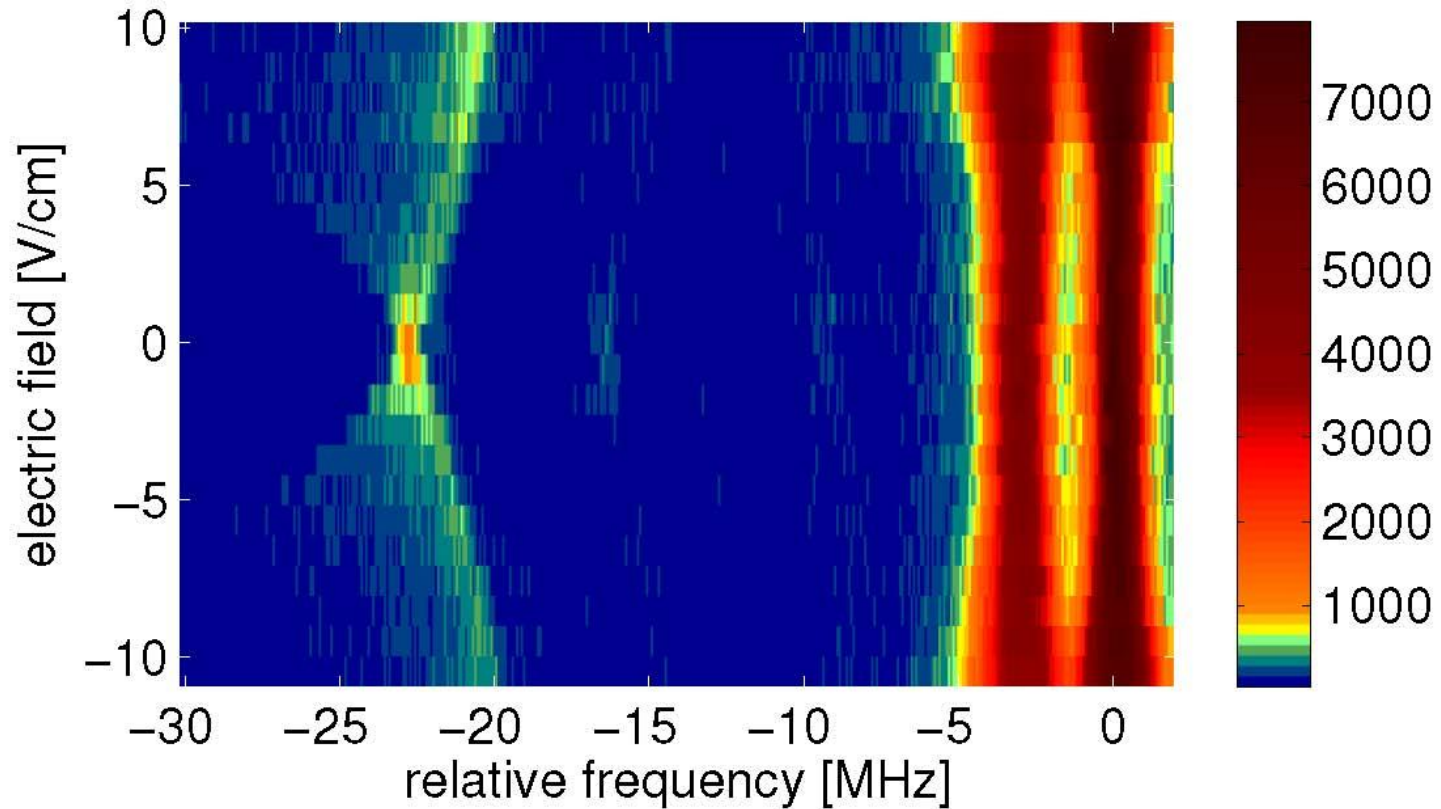
# Stark effect



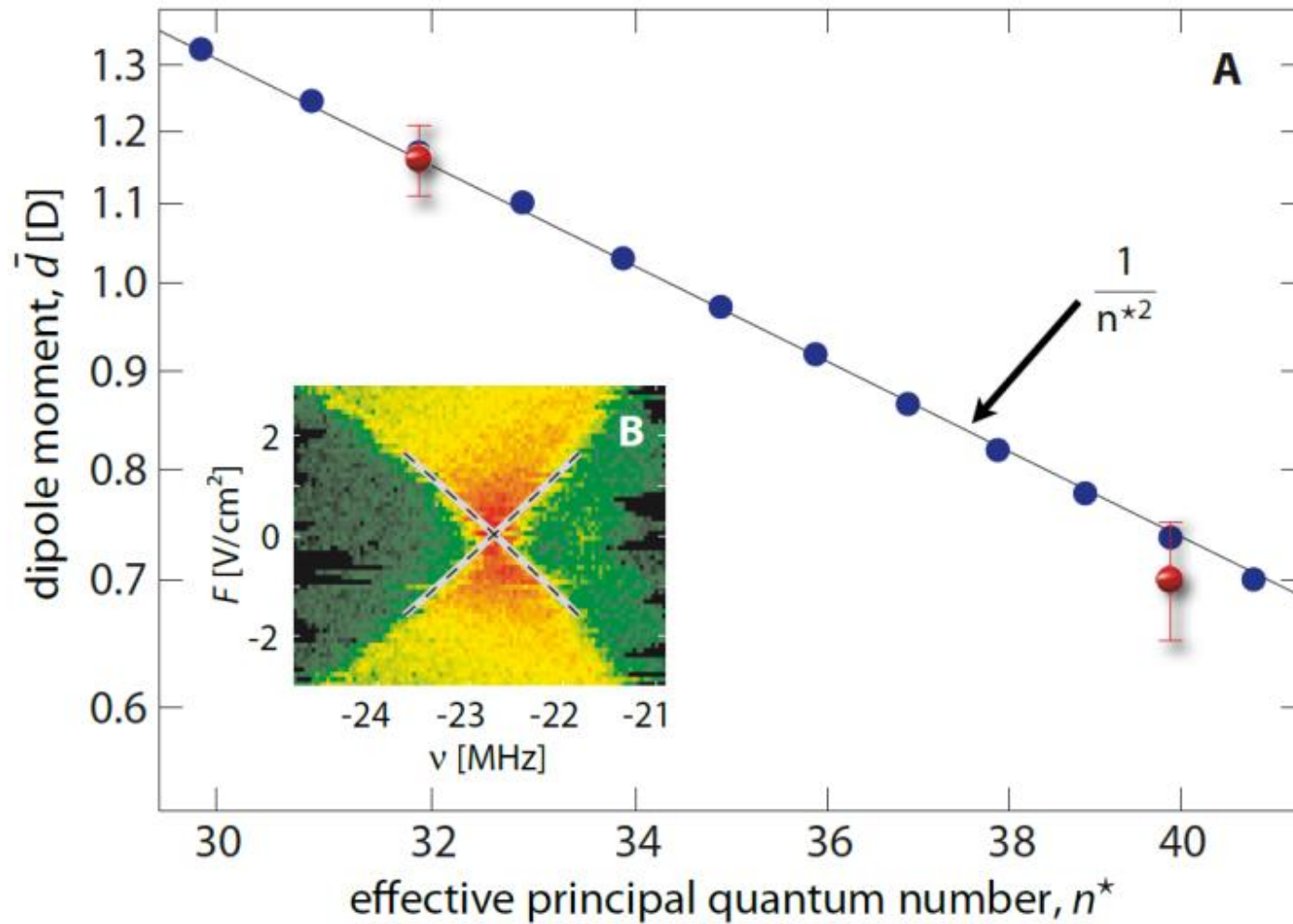
$$\Delta\nu = -\frac{1}{2}\alpha E^2 \quad \alpha = 4.19 \frac{\text{MHz}}{(\text{V/cm})^2}$$



# „Subtracted“ Stark effect

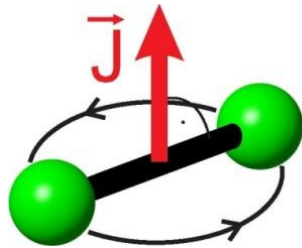








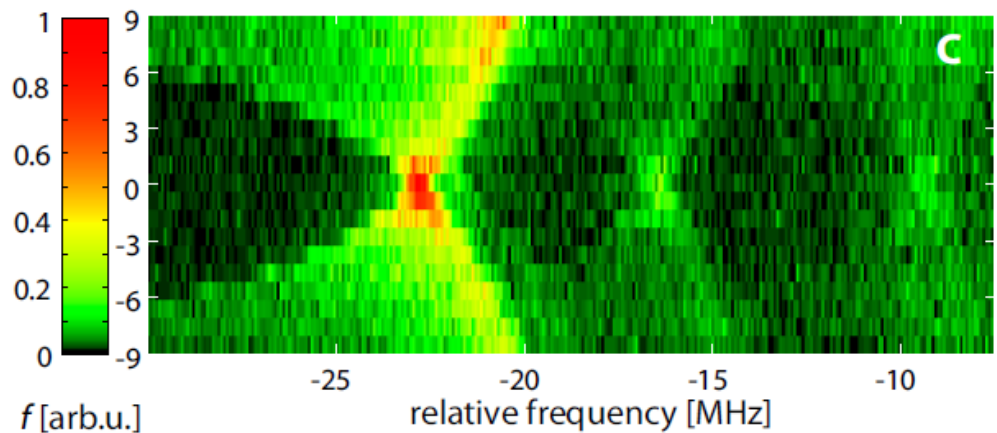
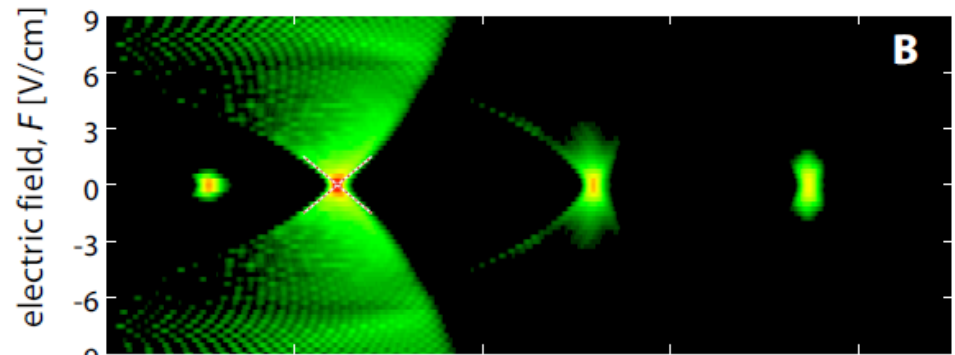
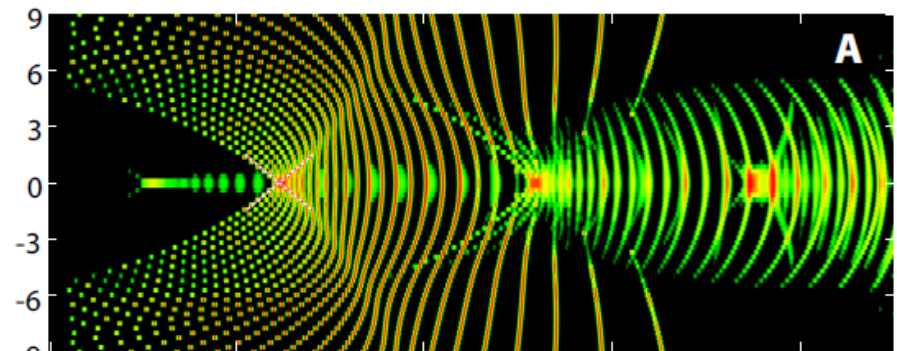
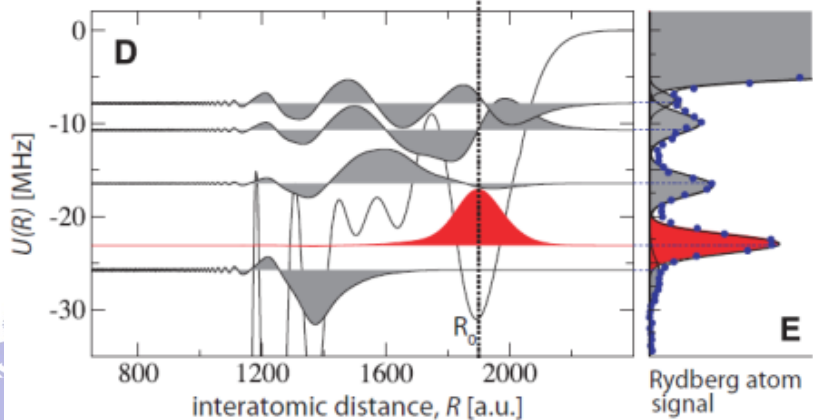
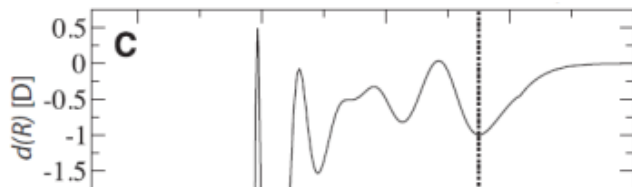
# Full theory



$B=11.5$  kHz

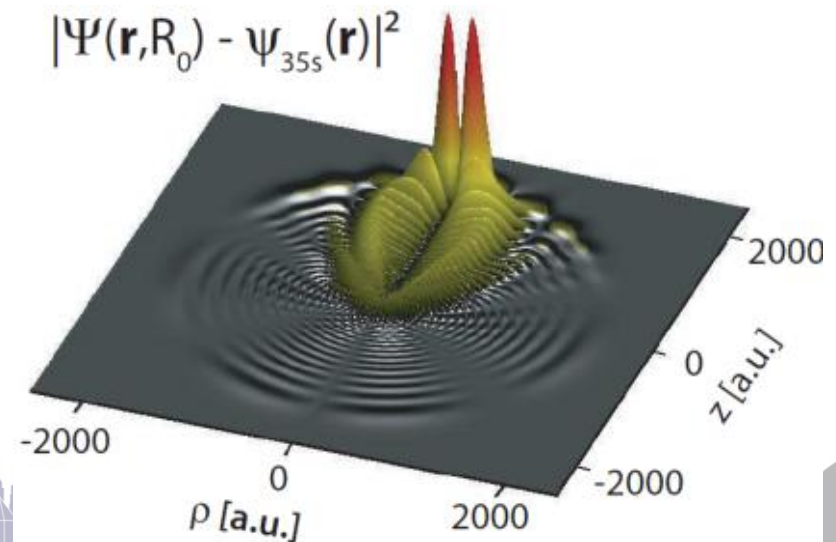
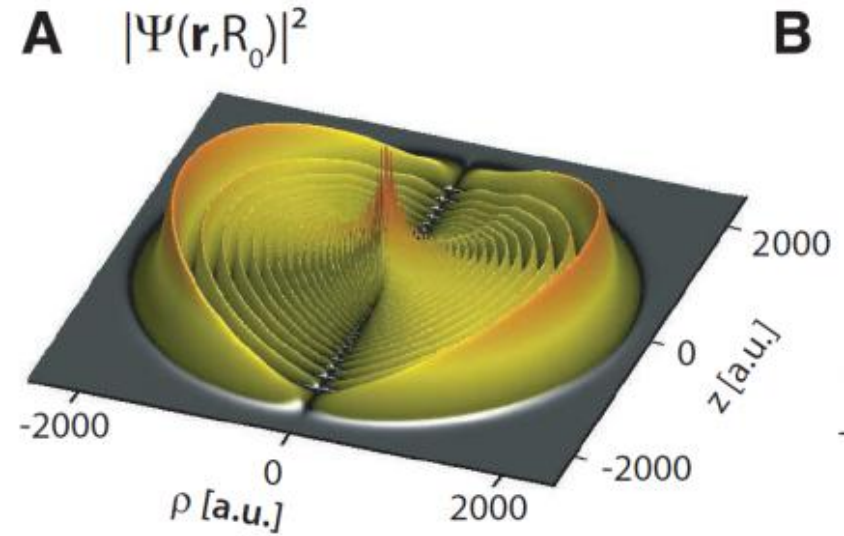
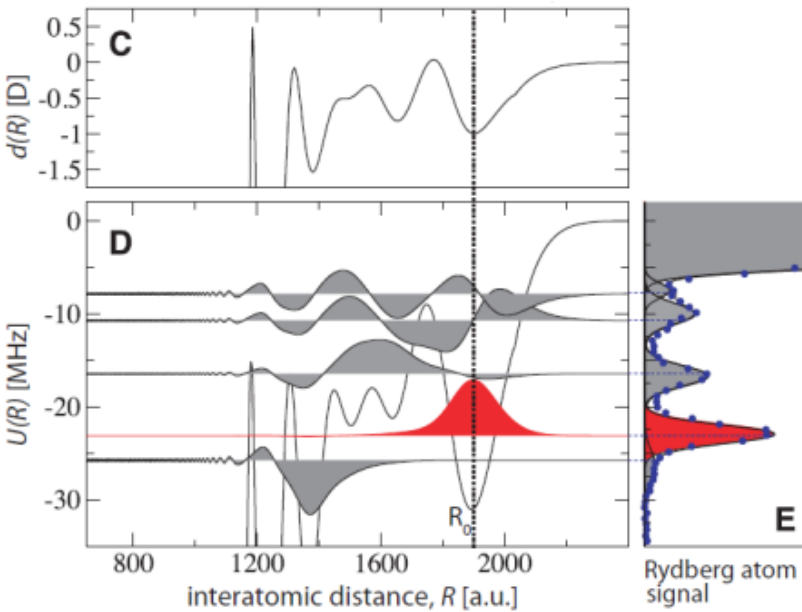
$T=3$   $\mu$ K

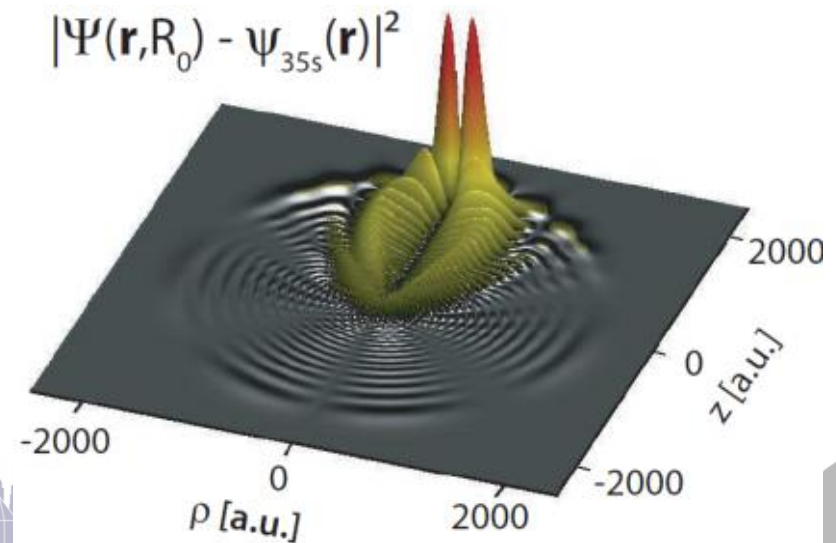
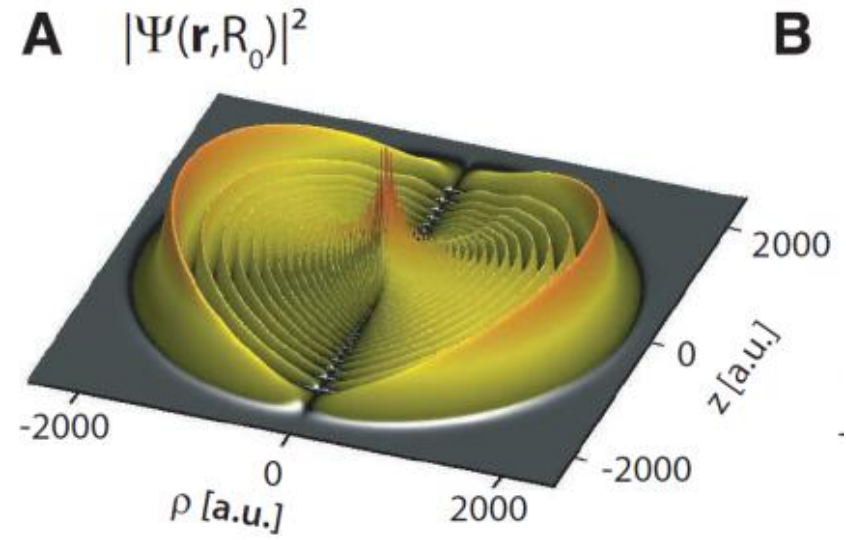
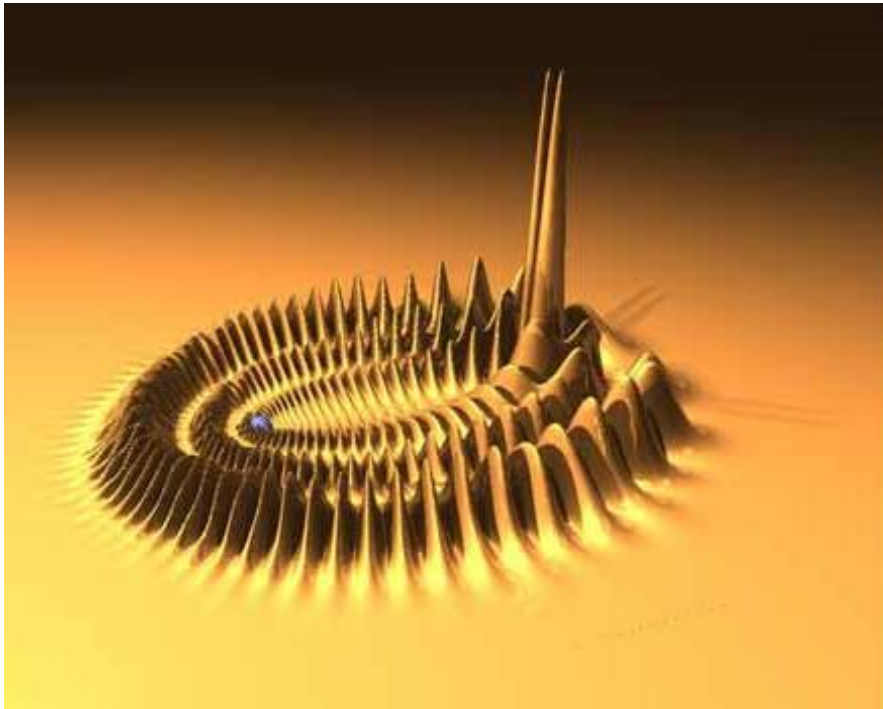
$\gamma=142$  kHz



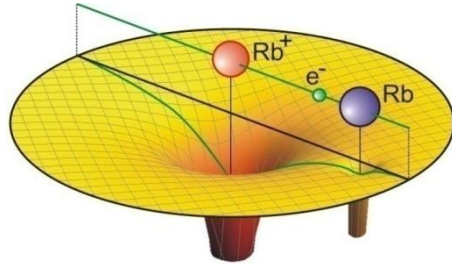
# Origin of the permanent dipole moment

$$d(R) = \langle \Psi(r, R) | ez | \Psi(r, R) \rangle$$

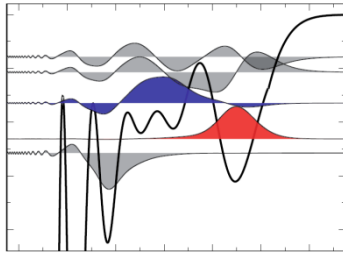




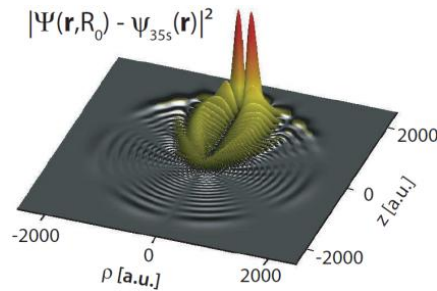
# Conclusion



**Ultra-longrange Rydberg Molecules**



**Decay mechanism of Rydberg molecules**

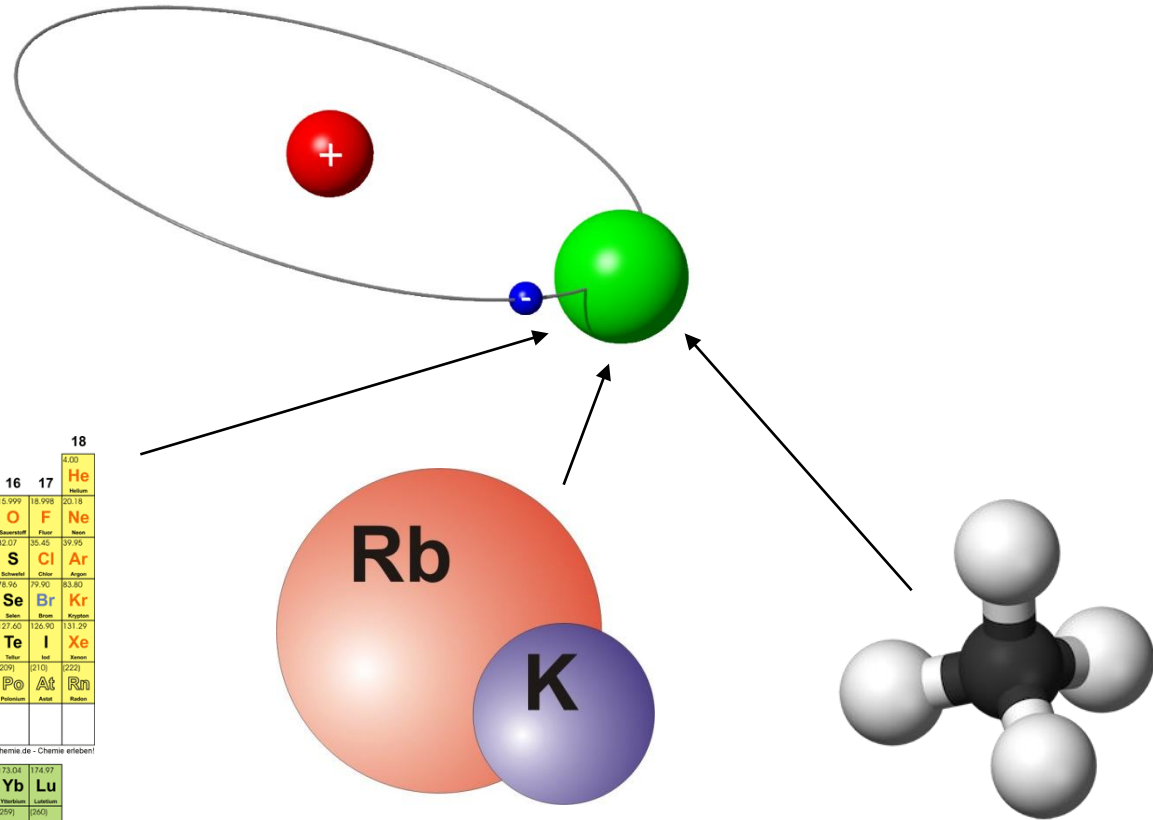


**Trilobite states**





# More exotic molecules ?



*Periodensystem der Elemente*

1	2	13	14	15	16	17	18																												
1 H 1,01 Wasserstoff	He 4,00 Helium	3 Li 6,94 Lithium	4 Be 9,01 Beryllium	5 B 10,81 Bor	6 C 12,01 Kohlenstoff	7 N 14,01 Stickstoff	8 O 15,99 Sauerstoff	9 F 18,99 Fluor	10 Ne 20,18 Neon																										
2 Na 22,99 Natrium	3 Mg 24,31 Magnesium	4 Al 26,98 Aluminium	5 Si 28,09 Silicium	6 P 30,97 Phosphor	7 S 32,07 Schwefel	8 Cl 35,45 Chlor	9 Ar 39,95 Argon	10 K 39,10 Kalium	11 Ca 40,08 Calcium	12 Sc 44,96 Scandium	13 Ti 47,88 Titan	14 V 50,94 Vanadium	15 Cr 52,00 Chrom	16 Mn 54,94 Mangan	17 Fe 55,85 Eisen	18 Co 58,93 Cobalt	19 Ni 58,71 Nickel	20 Cu 63,55 Kupfer	21 Zn 65,38 Zink	22 Ga 69,72 Gallium	23 Ge 72,61 Germanium	24 As 74,92 Arsen	25 Se 78,96 Selen	26 Br 79,90 Brom	27 Kr 83,80 Krypton										
3 Rb 85,47 Rubidium	4 Sr 87,52 Strontium	5 Y 88,91 Yttrium	6 Zr 91,22 Zirkon	7 Nb 92,91 Niob	8 Mo 95,94 Molybdän	9 Tc 98 Technetium	10 Ru 101,07 Ruthenium	11 Rh 102,91 Rhodium	12 Pd 106,42 Palladium	13 Ag 107,87 Silber	14 Cd 112,41 Cadmium	15 In 114,82 Indium	16 Sn 118,71 Zinn	17 Sb 121,76 Antimon	18 Te 127,60 Tellur	19 I 126,90 Jod	20 Xe 131,29 Xenon	21 Cs 132,91 Cäsium	22 Ba 137,33 Baryum	23 La-Lu 138,91 Lanthan	24 Hf 178,49 Hafnium	25 Ta 180,95 Tantal	26 W 183,84 Wolfram	27 Re 186,21 Rhenium	28 Os 190,23 Osmium	29 Ir 192,22 Iridium	30 Pt 195,08 Platin	31 Au 196,97 Gold	32 Hg 200,59 Quecksilber	33 Tl 204,38 Thallium	34 Pb 207,2 Blei	35 Bi 208,98 Bismut	36 Po 209 Polonium	37 At 210 Astat	38 Rn 222 Radon
4 Fr 223 Francium	5 Ra 226 Radium	6 Ac-Lr 227,03 Actinoid	7 Rf 261 Rutherfordium	8 Db 262 Dubnium	9 Sg 263 Seaborgium	10 Bh 264 Bohrium	11 Hs 265 Hassium	12 Mt 266 Meitnerium	13 Ds 269 Darmstadtium	14 La 138,91 Lanthan	15 Ce 140,12 Cer	16 Pr 140,91 Praseodym	17 Nd 144,24 Neodym	18 Pm 144,91 Promethium	19 Sm 150,36 Samarium	20 Eu 151,97 Europium	21 Gd 157,25 Gadolinium	22 Tb 158,93 Terbium	23 Dy 162,50 Dysprosium	24 Ho 164,93 Holmium	25 Er 167,26 Erbium	26 Tm 168,93 Thulium	27 Yb 173,04 Ytterbium	28 Lu 174,97 Lutetium											
5 Ac 227,03 Actinoid	6 Th 232,04 Thorium	7 Pa 231,04 Protactinium	8 U 238,03 Uran	9 Np 237 Neptunium	10 Pu 244 Plutonium	11 Am 243 Americium	12 Cm 247 Curium	13 Bk 247 Berkelium	14 Cf 251 Californium	15 Es 252 Einsteinium	16 Fm 257 Fermium	17 Md 258 Mendelevium	18 No 259 Nobelium	19 Lr 260 Lawrencium																					

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# Radial dynamics

