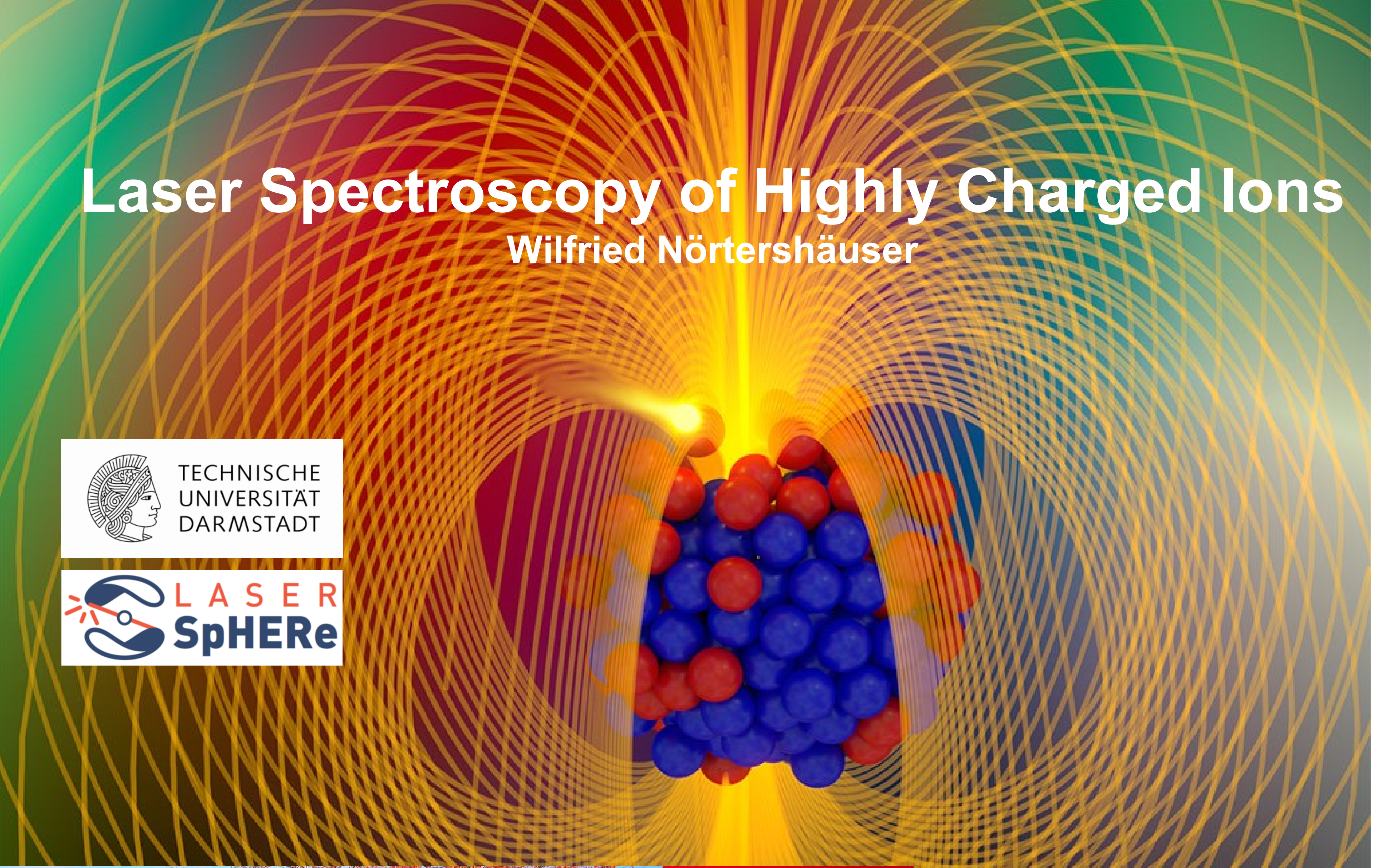


Laser Spectroscopy of Highly Charged Ions

Wilfried Nörtershäuser

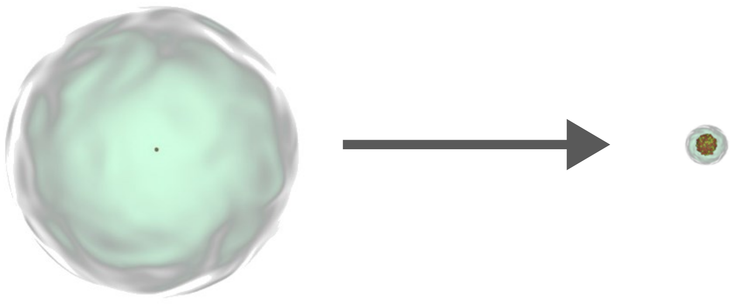
TECHNISCHE
UNIVERSITÄT
DARMSTADT

LASER
SpHERe

DE LA RECHERCHE À L'INDUSTRIE
cea
ESNT
Espace de Structure Nucléaire Théorique

Workshop
"Laser spectroscopy as a tool for nuclear theories"

Interest in Highly Charged Ions (HCI)



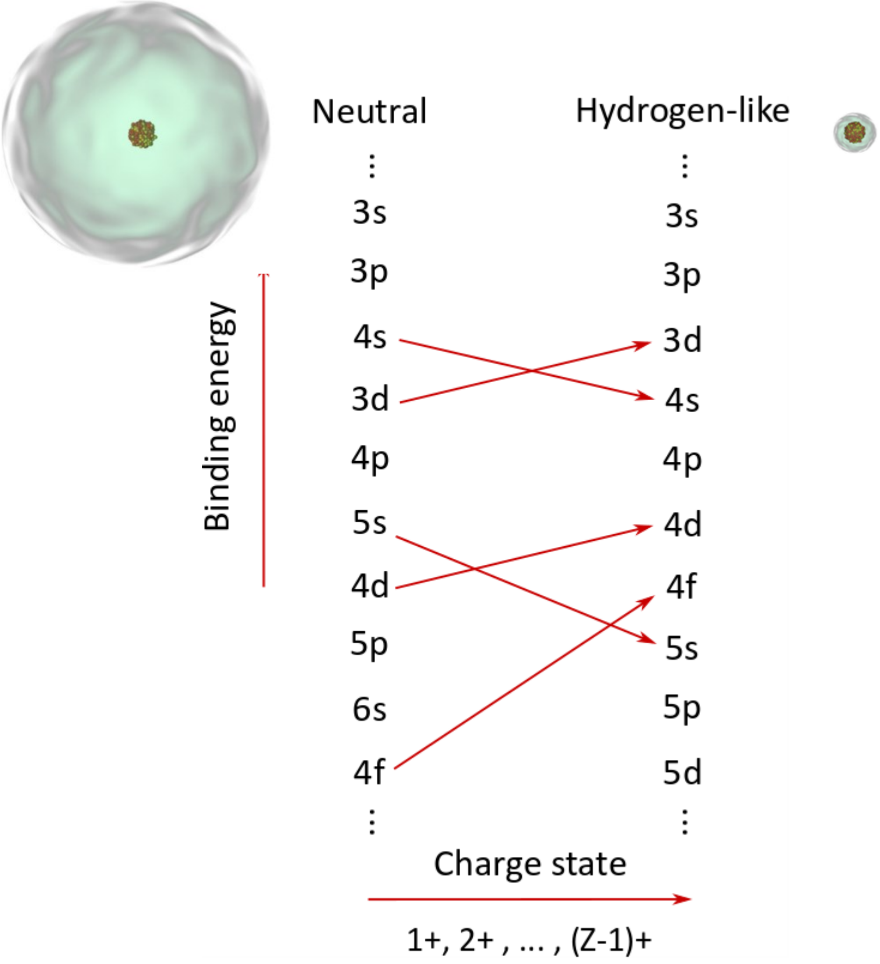
Radius	Z^{-1}
Polarizability	Z^{-1}
Gross structure	Z^{2}
Fine structure splitting	Z^{4}
Hyperfine structure	Z^{3}
QED effects	Z^{4}

- Relativistic, QED, nuclear effects
- Forbidden optical transitions sensitive to $\dot{\alpha}, \dot{\mu}$, violation of local Lorentz invariance

Kozlov et al., Rev. Mod. Phys. **90**, 045005 (2018) , arXiv:180306532

Courtesy of Peter Micke, PTB

Optical Transitions in HCI



Neutral

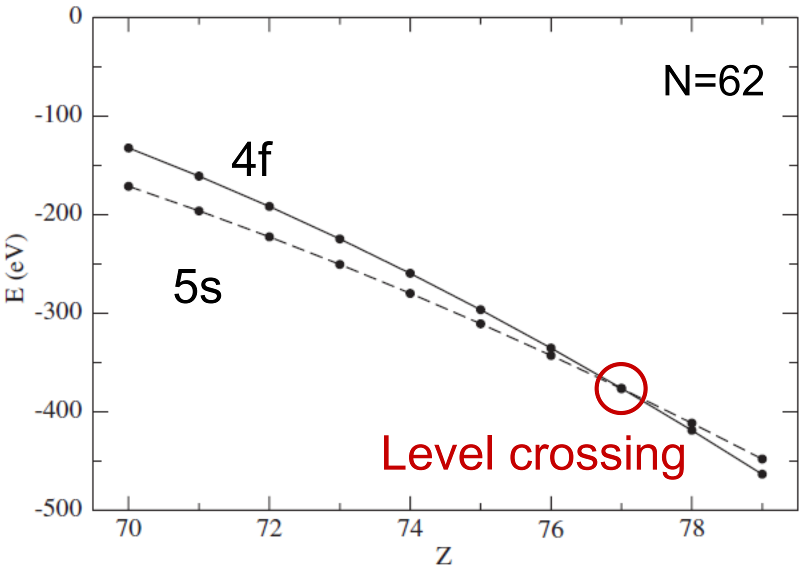
Hydrogen-like

Binding energy

Neutral	Hydrogen-like
⋮	⋮
3s	3s
3p	3p
4s	3d
3d	4s
4p	4p
5s	4d
4d	4f
5p	5s
6s	5p
4f	5d
⋮	⋮

Charge state

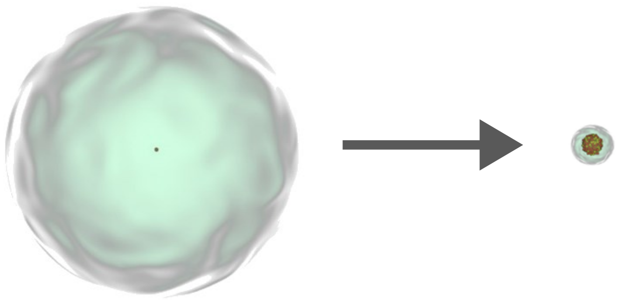
1+, 2+ , … , (Z-1)+



Berengut et al., PRL **106**, 210802 (2011)

Courtesy of Peter Micke, PTB

Advantages of HCI for High-Precision Experimens and Optical Clocks



Linear Stark shift	Z^{-1}
Second order Stark shift	Z^{-4}
Linear Zeeman shift	Z^{0}
Second order Zeeman shift	$Z^{-3...-4}$
Electric quadrupole shift	Z^{-2}

Less sensitive to external perturbations

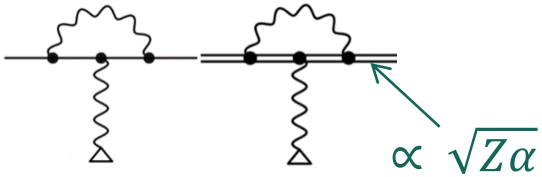
Courtesy of Peter Micke, PTB

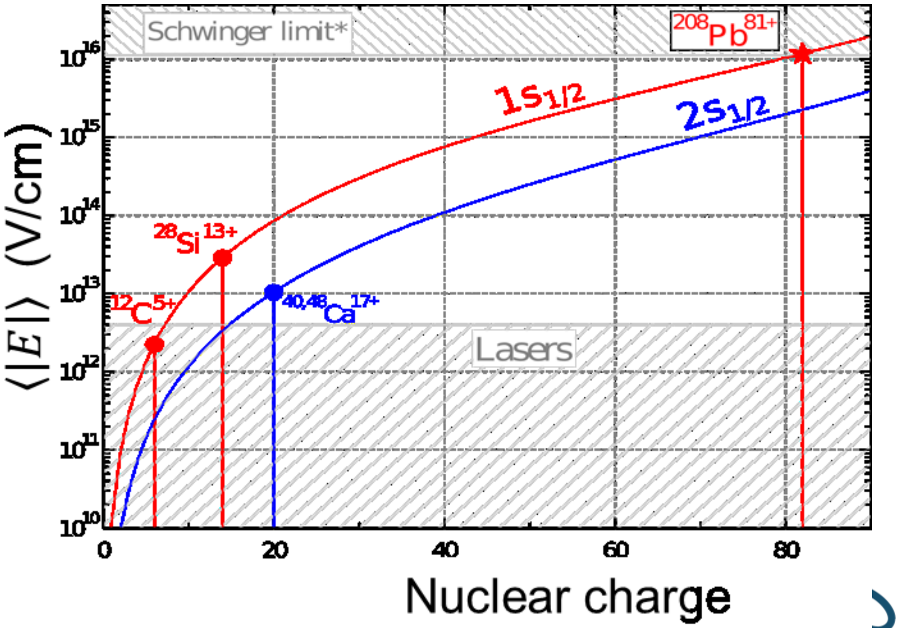
QED in Highly Charged Ions (HCI)

- Impressive theory predictions and most precise experimental results, e.g. electron magnetic moments: *g*-2, *g*-factor in HCI [1]
  → Most stringently tested theory in weak fields

- Validity of QED in strong fields?
  Test of bound state QED (BS-QED) under extreme conditions in high electric and magnetic fields of heavy HCI
  - *g*-factor measurements in H or Li-like ions
  - Fine structure and hyperfine structure spectroscopy

  → Combine strongest field and highest precision





[1] H. Häffner, et al., Phys. Rev. Lett. 85, 5308 (2000)
J.Verdu, et al., Phys. Rev. Lett. 92, 093002 (2004)
[2] S. Sturm, et al., Nature 506, 7489 (2014)
[3] S. Sturm, et al., Phys. Rev. Lett. 107, 023002 (2011)
[4] F. Köhler, et al. Nat. Comm. 7, 10246 (2016)

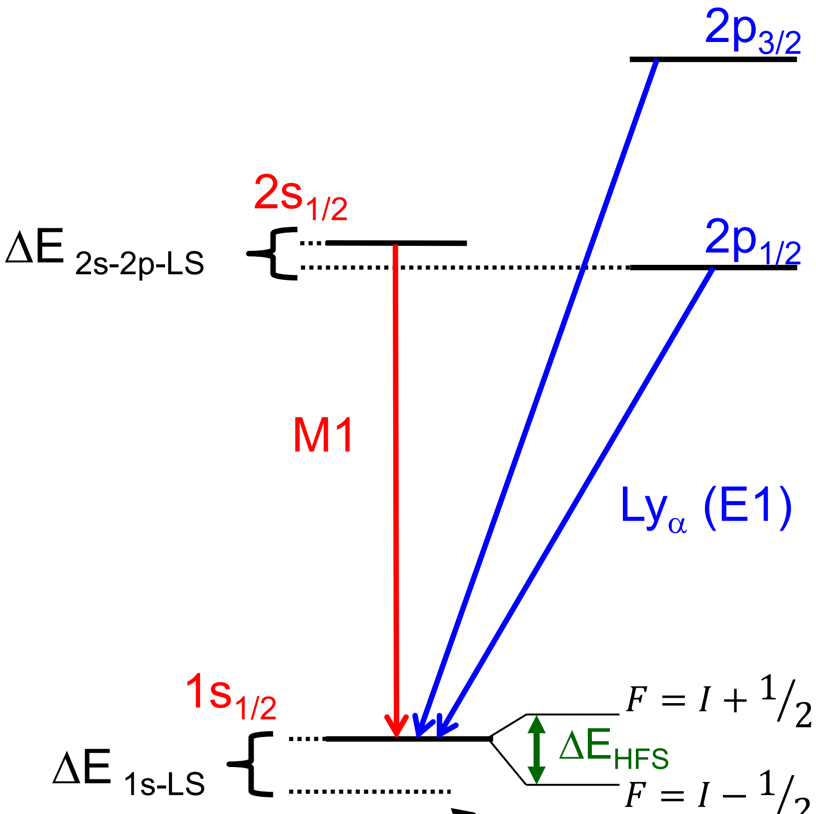
Outline

Laser Spectroscopy in Heavy Highly Charged Ions and the „Hyperfine Puzzle of strong-field bound-state QED“

Nuclear Magnetic Resonance and ist contribution to a solution of the „Hyperfine Puzzle“

Into the future: Experimental developments towards high-precision spectroscopy in HCI

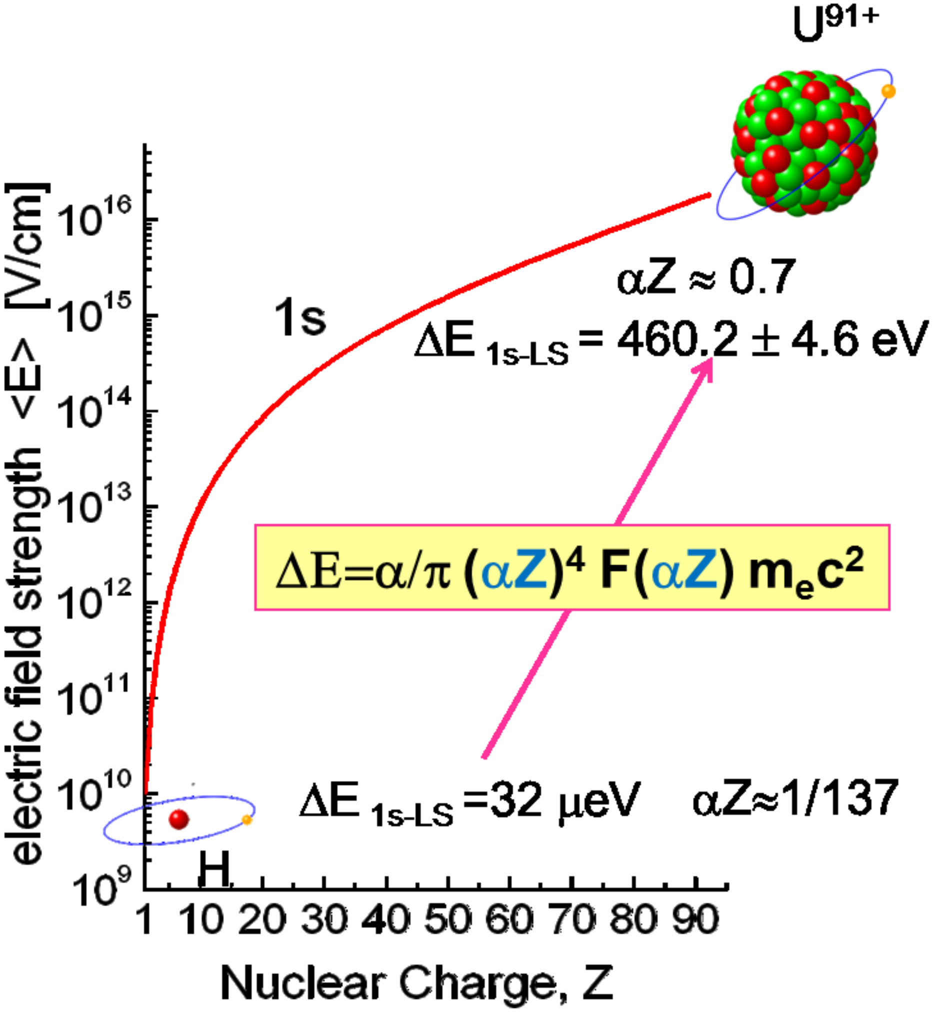
Structure of Heavy H-Like Ions



Quantum
Electro-
Dynamics

$$(i\gamma^\mu\partial_\mu - m)\psi = 0$$

Dirac Equation

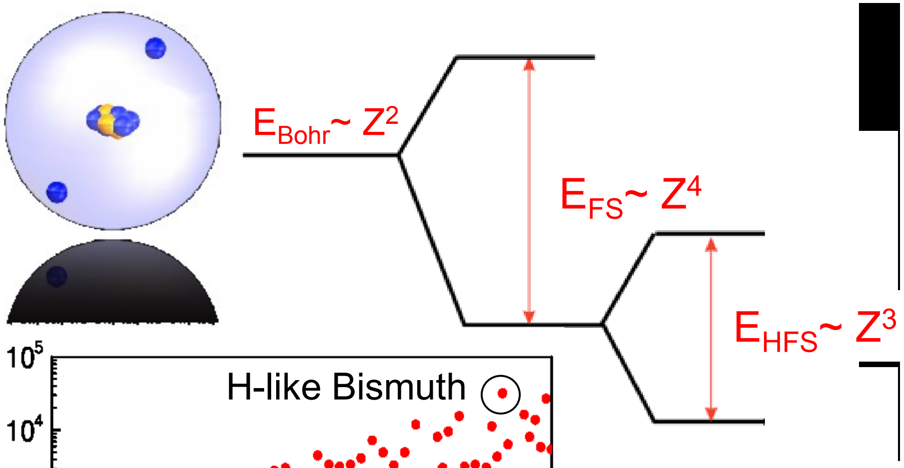


$$\Delta E = \alpha/\pi \, (\alpha Z)^4 \, F(\alpha Z) \, m_e c^2$$

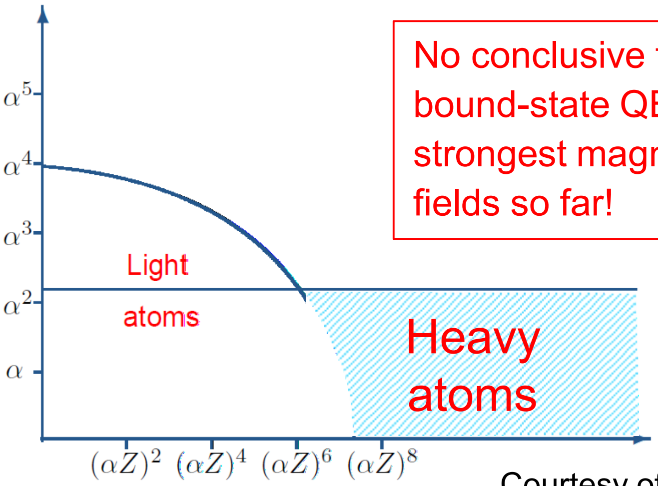
A. Gumberidze et al., PRL 94, 223001 (2005)

Test of BS-QED in Highly Charged Ions

atomic structure:

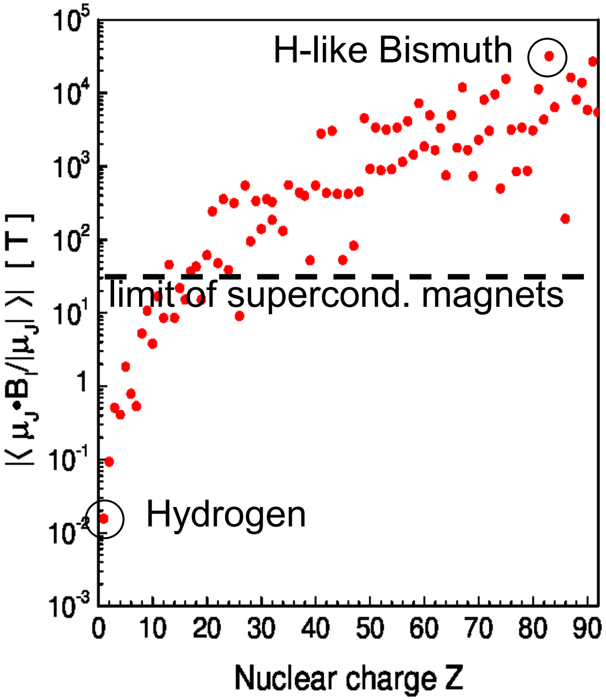


Tests of QED to lowest orders in α and to all orders in αZ

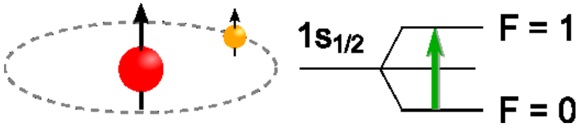


No conclusive test of bound-state QED in the strongest magnetic fields so far!

Courtesy of V. Shabaev

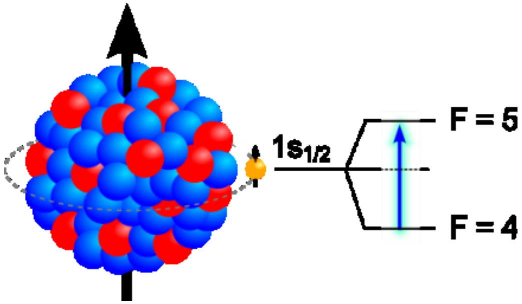


H: λ = 21 cm
τ = 11 Ma

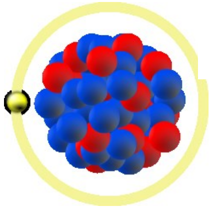


$^{209}Bi^{82+}$: λ = 244 nm
τ = 0.39 ms

$^{209}Bi^{80+}$: λ = 1555 nm
τ = 83.3 ms



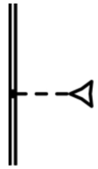
HFS-Measurements in Heavy H-Like Ions



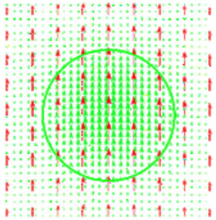
Bi^{82+}

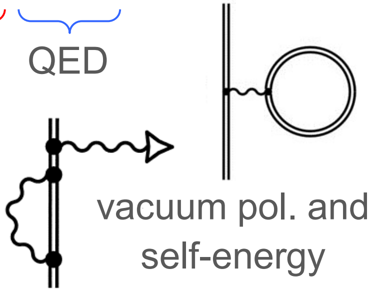
$$\Delta E^{(1s)} = \underbrace{\frac{4}{3}\alpha(\alpha Z)^3 \frac{\mu_I}{\mu_N}\frac{m}{m_p}\frac{2I+1}{2I} mc^2 \times \big(A(\alpha Z)^{1s}}_{\text{Dirac}} \underbrace{(1-\epsilon^{1s}_{BR})(1-\epsilon^{1s}_{BW})}_{\text{nuclear contributions}} + \underbrace{x^{1s}_{red}}_{\text{QED}}\big)$$

known (?)

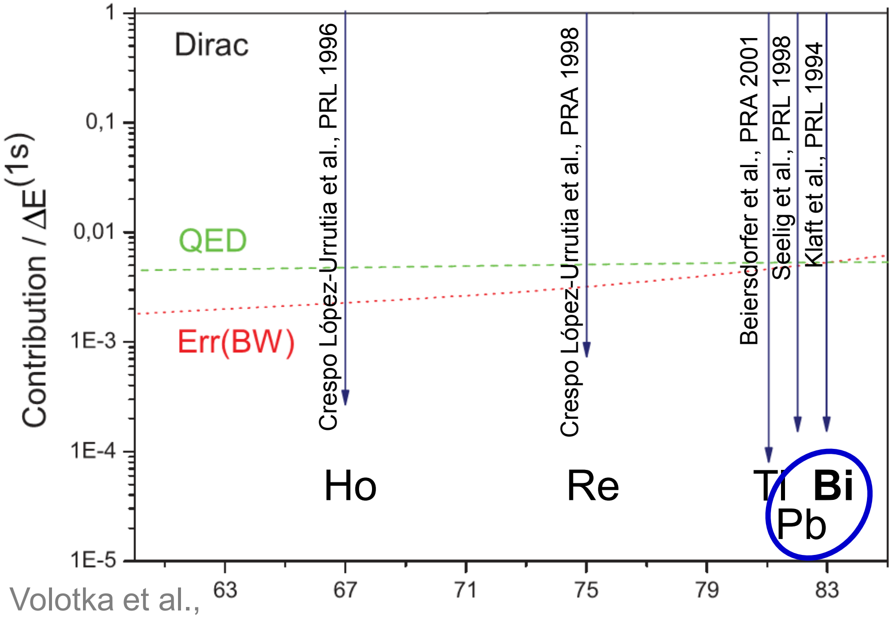


Bohr-Weisskopf effect
Breit-Rosenthal effect
nuclear polarization

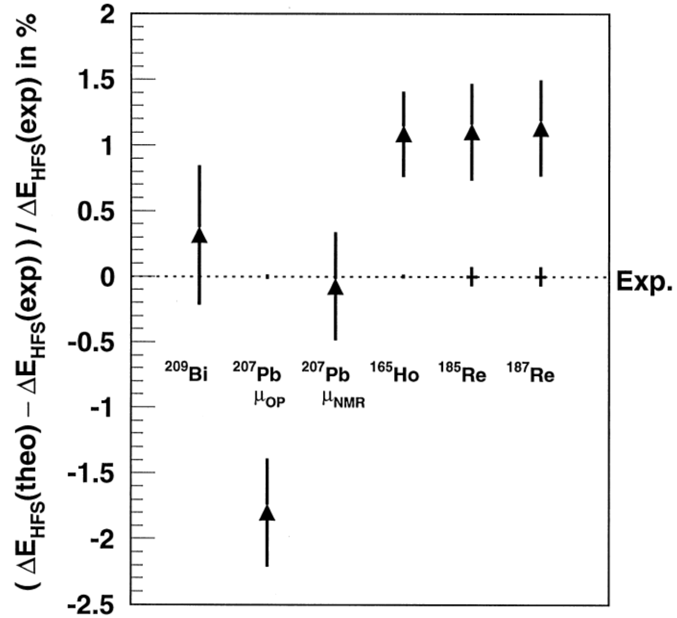




vacuum pol. and
self-energy

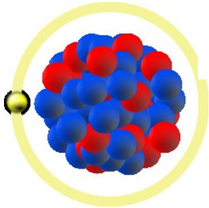


A.V. Volotka et al.,
Ann. Physik 525, 636 (2013)



T.Beier, Phys.Rep. **339**, 79 (2000)

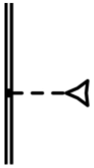
Theory: Trouble & Solution



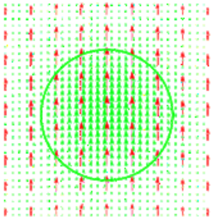
Bi^{82+}

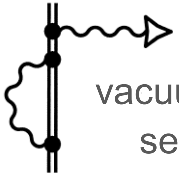
$$\Delta E^{(1s)} = \underbrace{\frac{4}{3}\alpha(\alpha Z)^3 \frac{\mu_I}{\mu_N}\frac{m}{m_p}\frac{2I+1}{2I}mc^2 \times}_{\text{Dirac}} \big(\underbrace{A(\alpha Z)^{1s}(1-\epsilon_{BR}^{1s})(1-\epsilon_{BW}^{1s})}_{\text{nuclear contributions}} + \underbrace{x_{red}^{1s}}_{\text{QED}}\big)$$

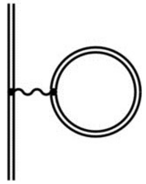
known (?)



Bohr-Weisskopf effect
Breit-Rosenthal effect
nuclear polarization

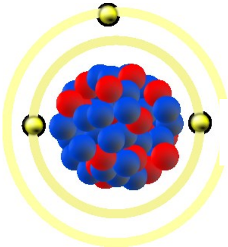






vacuum pol. and self-energy

uncertainty Bohr-Weisskopf > QED contribution



Bi^{80+}

$$\Delta E^{(2s)} = \frac{1}{6}\alpha(\alpha Z)^3 \frac{\mu_I}{\mu_N}\frac{m}{m_p}\frac{2I+1}{2I}mc^2 \times \big(A(\alpha Z)^{2s}(1-\epsilon_{BR}^{2s})(1-\epsilon_{BW}^{2s}) + x_{red}^{2s} + B_Z + C_Z\big)$$

specific difference (V. M. Shabaev et al. PRL 86 (2001))

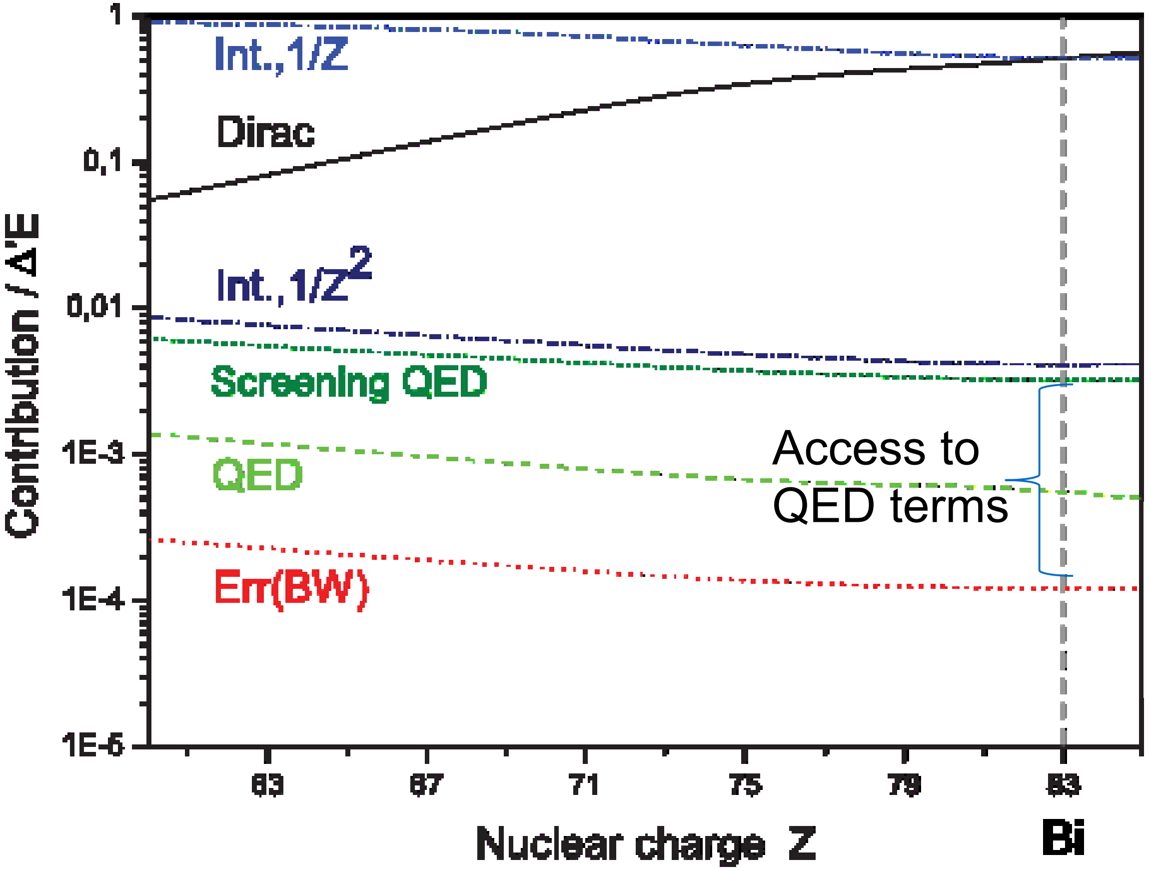
$$\boxed{\Delta' E = \Delta E^{(2s)} - \xi\ \Delta E^{(1s)}}$$

nuclear contributions removed

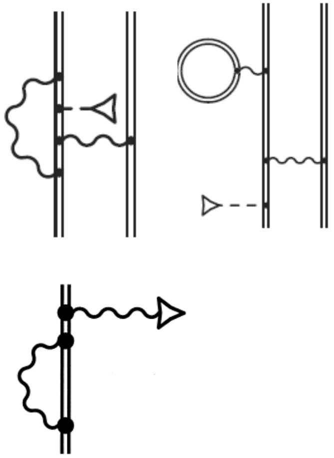
Precision Test of QED: The Specific Difference

specific difference (V. M. Shabaev et al. PRL 86 (2001)

$$\Delta' E = \Delta E^{(2s)} - \xi \, \Delta E^{(1s)}$$

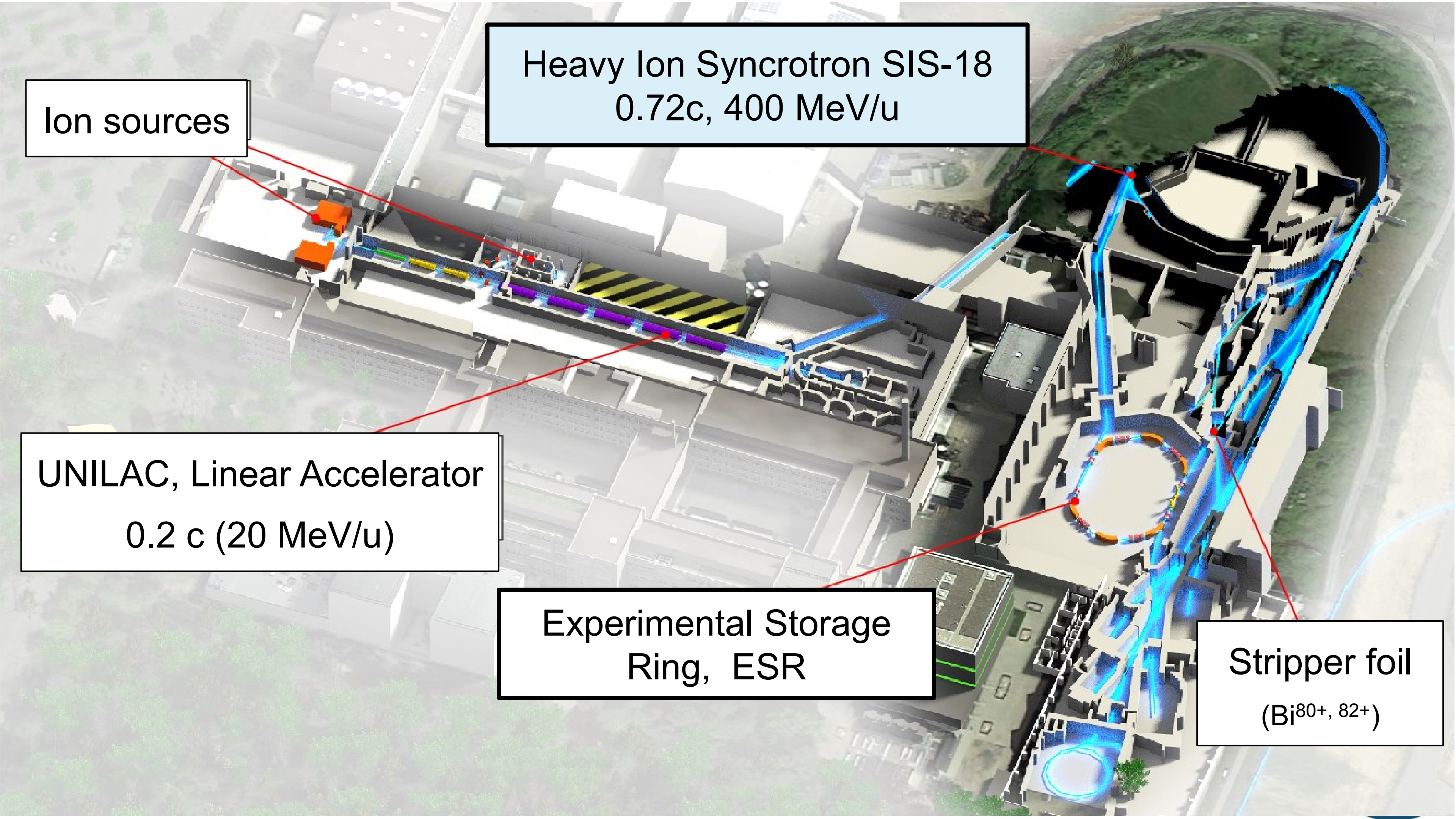


Access to QED terms



A. Volotka et al., Ann. Phys. **525**, 636 (2013)

Production of $^{209}\mathrm{Bi}^{80,82+}$



Ion sources

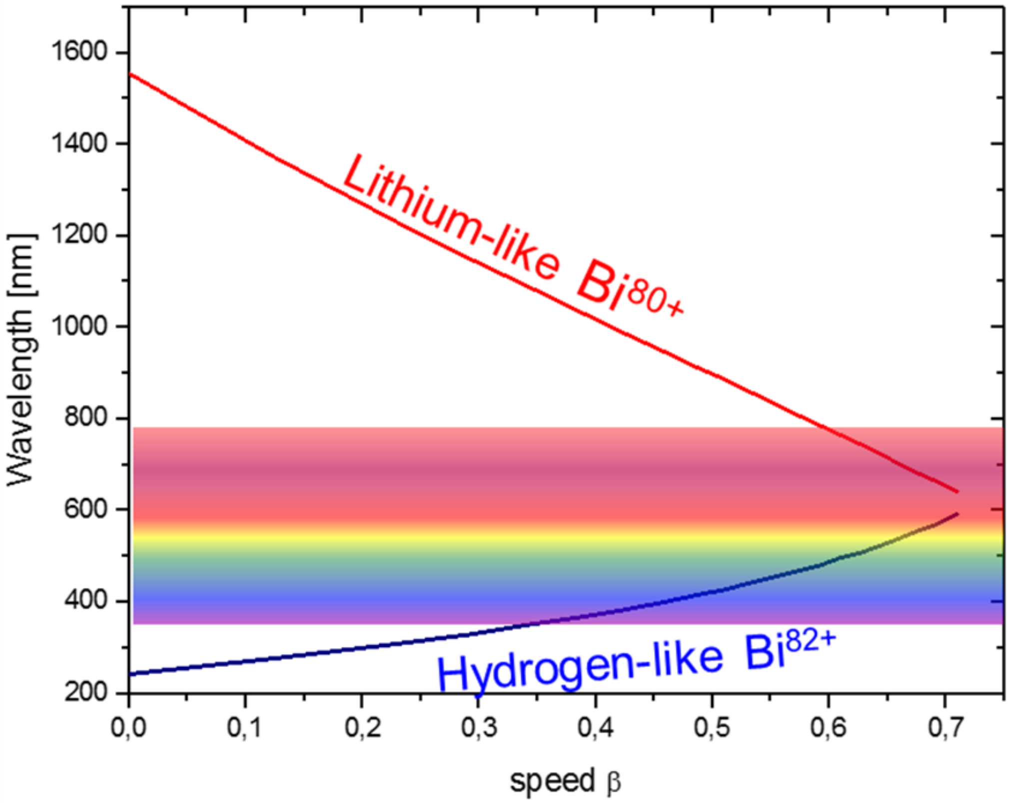
Heavy Ion Syncrotron SIS-18
0.72c, 400 MeV/u

UNILAC, Linear Accelerator
0.2 c (20 MeV/u)

Experimental Storage Ring, ESR

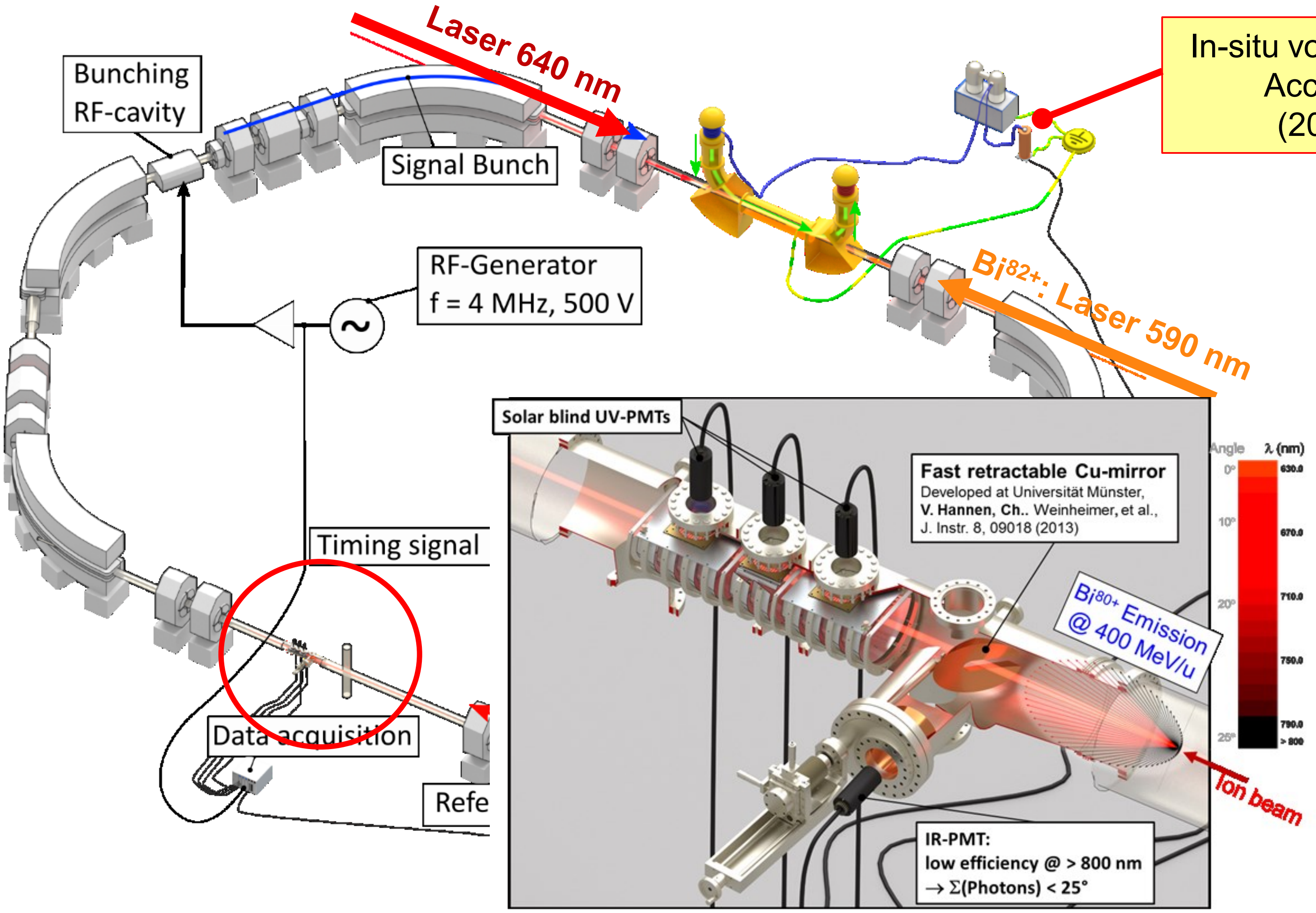
Stripper foil
($\mathrm{Bi}^{80+,\ 82+}$)

Reminder: Optical Doppler Effect



… for Dummies !

First High Accuracy Measurement of Δ'E



Fast retractable Cu-mirror
Developed at Universität Münster,
V. Hannen, Ch.. Weinheimer, et al.,
J. Instr. 8, 09018 (2013)

In-situ voltage measurement
Accuracy: ΔU~ 4V
(2011: ΔU=110V)

The Experimental Storage Ring ESR

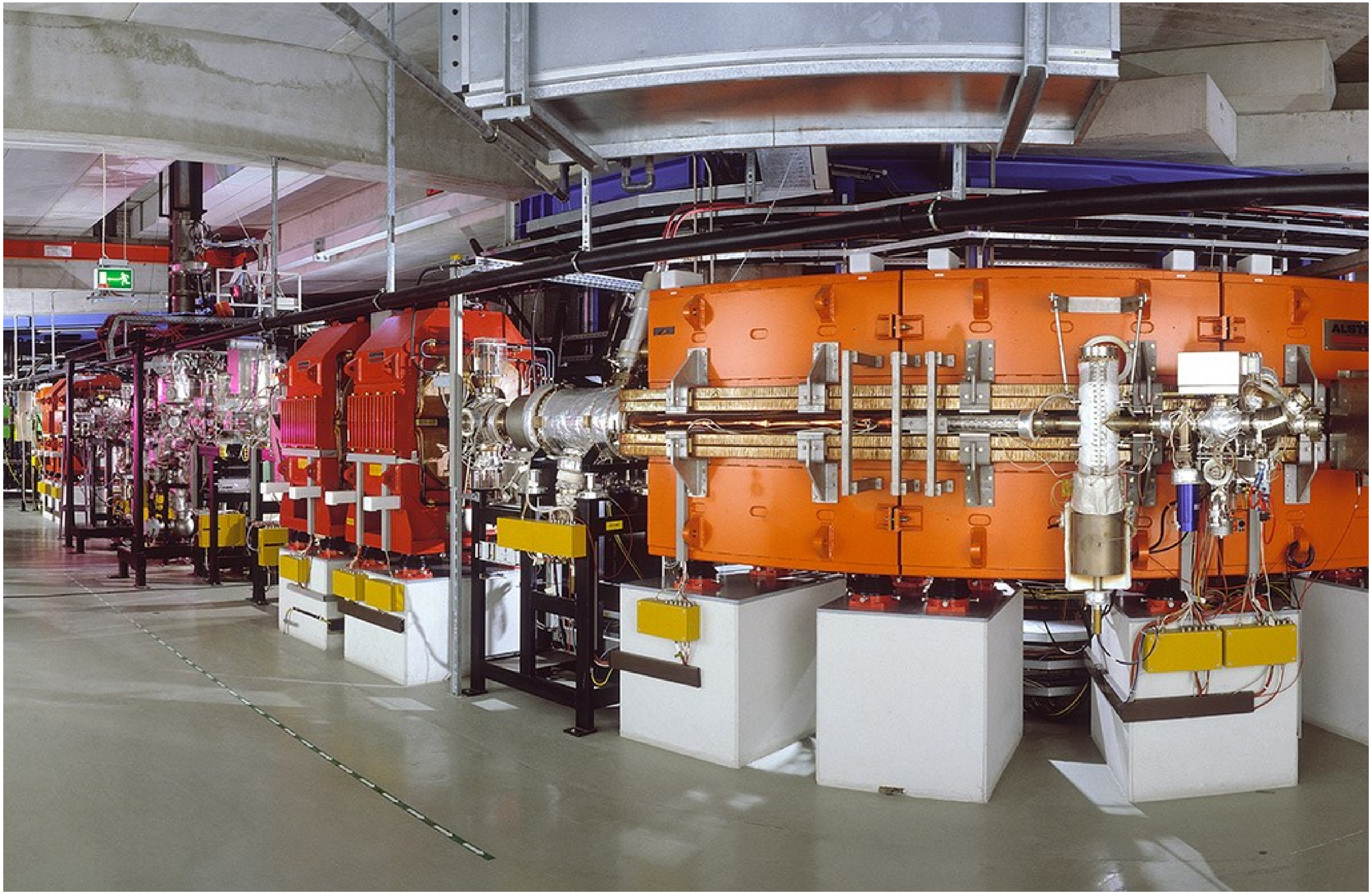
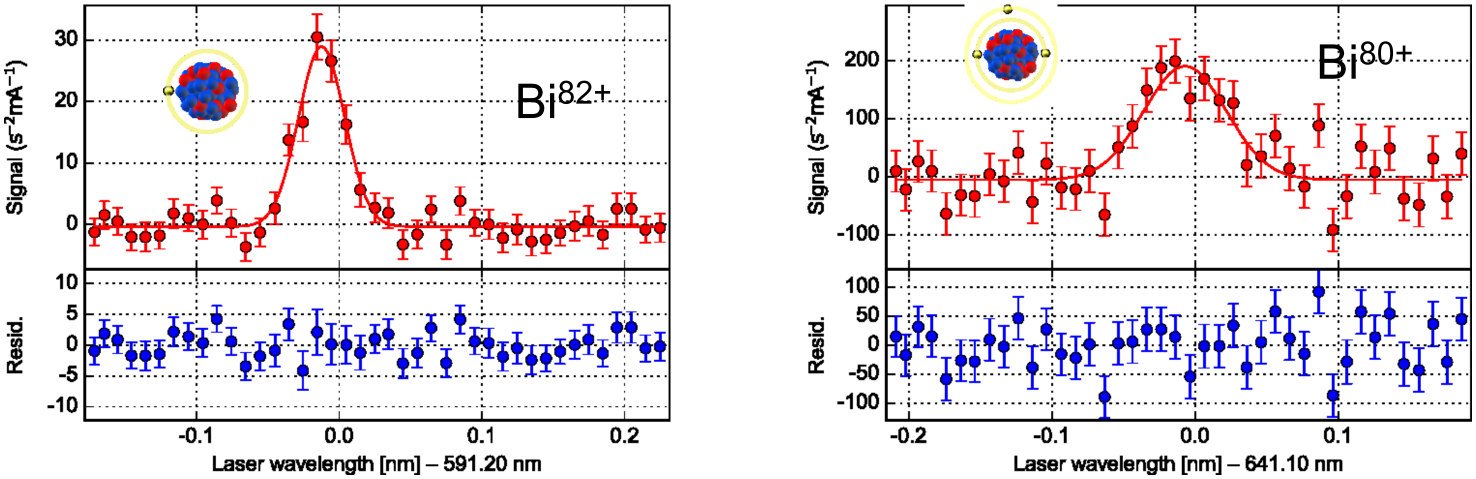
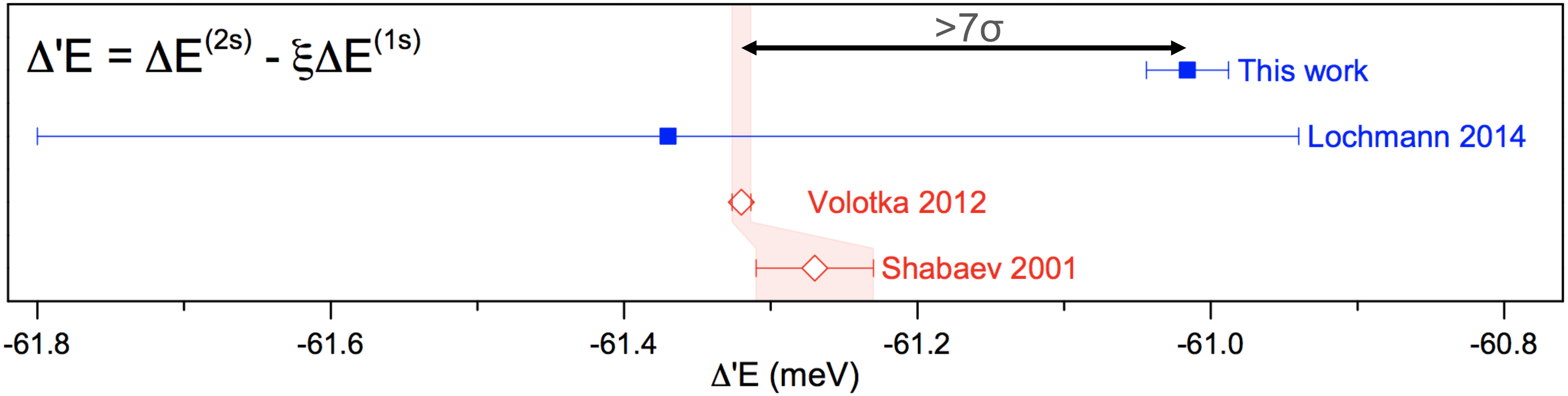


Foto:
J. Mai,
GSI

Results: Hyperfine Transition Wavelength





J. Ullmann et al., Nature Comm. 8, 15484 (2017)

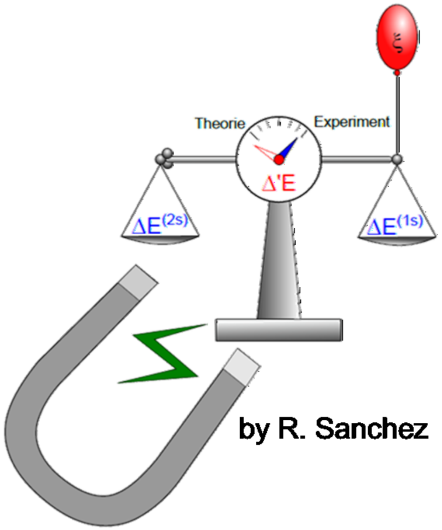
Explanations for the Discrepancy ?

Experiment

- ~~Our experimental value is wrong?!~~

unlikely

- The literature value of the nuclear magnetic moment of ^{209}Bi is wrong?!
  (Dirac term of the ground state hfs is proportional to the experimental value)



by R. Sanchez

Theory

- The specific difference does not work as expected?!

$$\Delta' E = \Delta E^{(2s)} - \xi \, \Delta E^{(1s)}$$

- ~~BS Strong-Field QED is wrong?!~~

First exclude other possibilities !

Challenge: Magnetic Moments Required

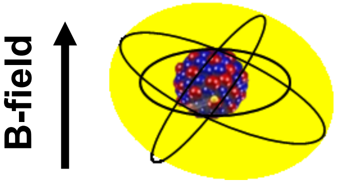
Author	$(1-\sigma_{\text{dia}})^{-1}$	σ_{chem}	$\mu(^{209}\text{Bi})\ [\mu_N]$
Proctor & Yu[1] [27]			4,0400(7)[3]
Ting & Williams[1] [28]	1,0104		4,0810(4)
Flynn[1] [31]			4,0391(2)[3]
Raghavan[2] [32]	1,0177		4,1106(2)
Baştuğ et al.[2] [33]	1,01757(6)		4,1103(5)
Gustavsson & Mårtensson-Pendrill[3] [30]		0,000(1)	4,110(4)

$Bi(NO_3)_3\,5H_2O$ solved in HNO_3 (nitric acid)

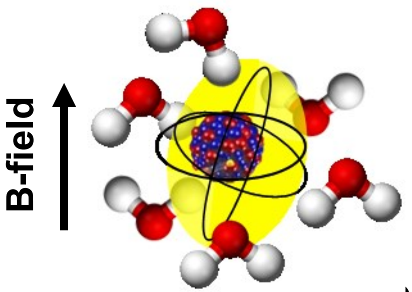
$$\mu_{I,\text{Exp}} = \frac{\Delta' E_{\text{Exp}}}{\Delta' E_{\text{Theo}}} \cdot \mu_{I,\text{Lit}}$$

$$\mu_{\text{Probe}} = \mu'_{\text{Probe}} / [1 - (\sigma_{\text{dia}} + \sigma_{\text{chem}})]$$

diamagnetic shielding

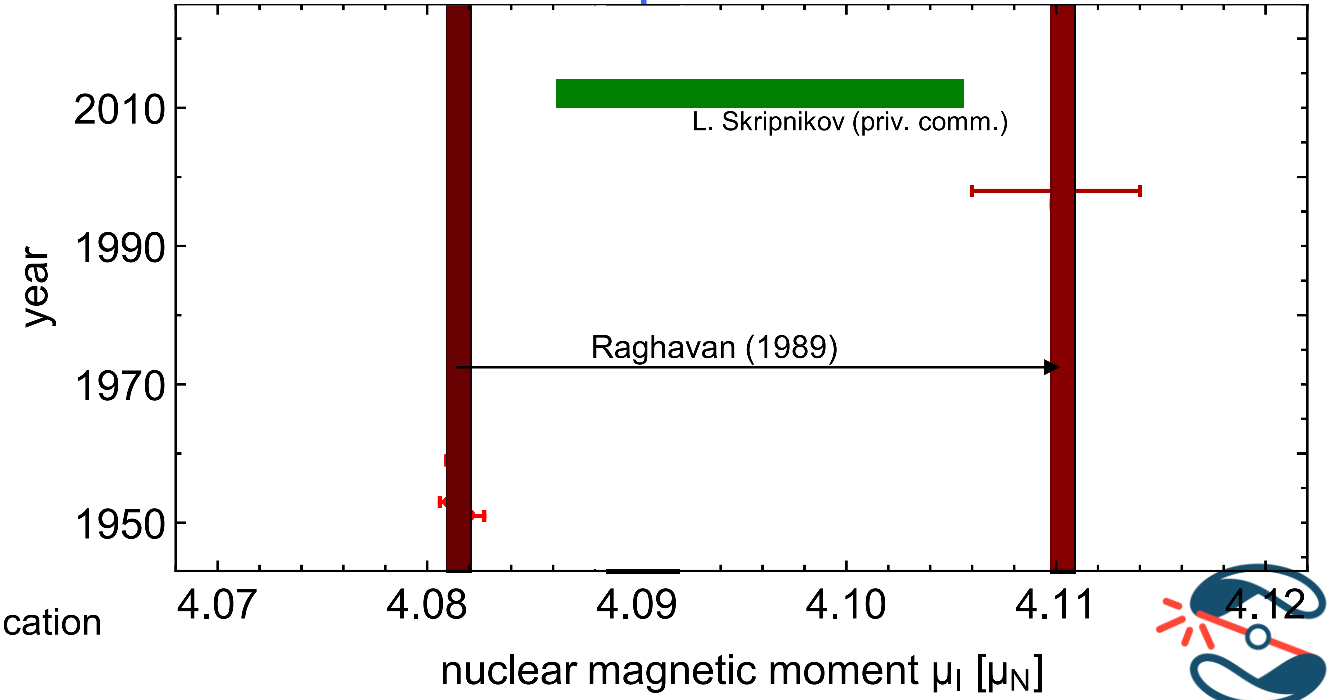


chemical shift

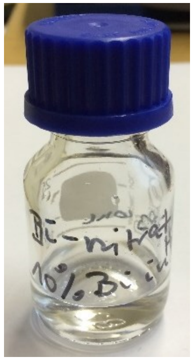


$[Bi(H_2O)_6]^{3+}$ cation

[Fedotov, M. A.; et al.
Neorg. Khim. 1998, 43, 307–310]



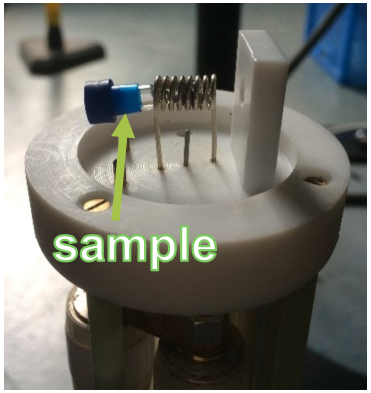
NMR Measurements @ TU Darmstadt



sample

$Bi(NO_3)_3$ solved
HNO_3 (nitric acid)

samples prepared
at Institut für Kernchemie
(Uni Mainz)



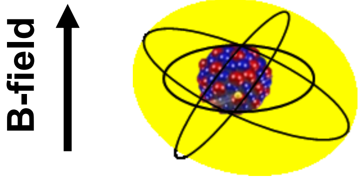
Cooperation: Prof. Michael Vogel

TU Darmstadt, Institut für Festkörperphysik

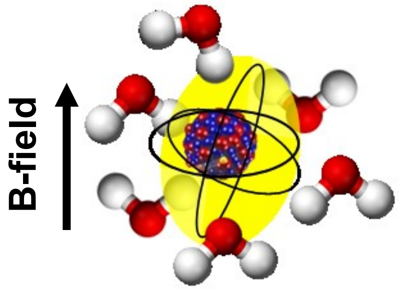
Chemical Shift in NMR Spectra of $Bi(NO_3)_{3\,(aq)}$

$$\mu_{\text{Probe}} = \mu'_{\text{Probe}}/[1-(\sigma_{\text{dia}}+\sigma_{\text{chem}})]$$

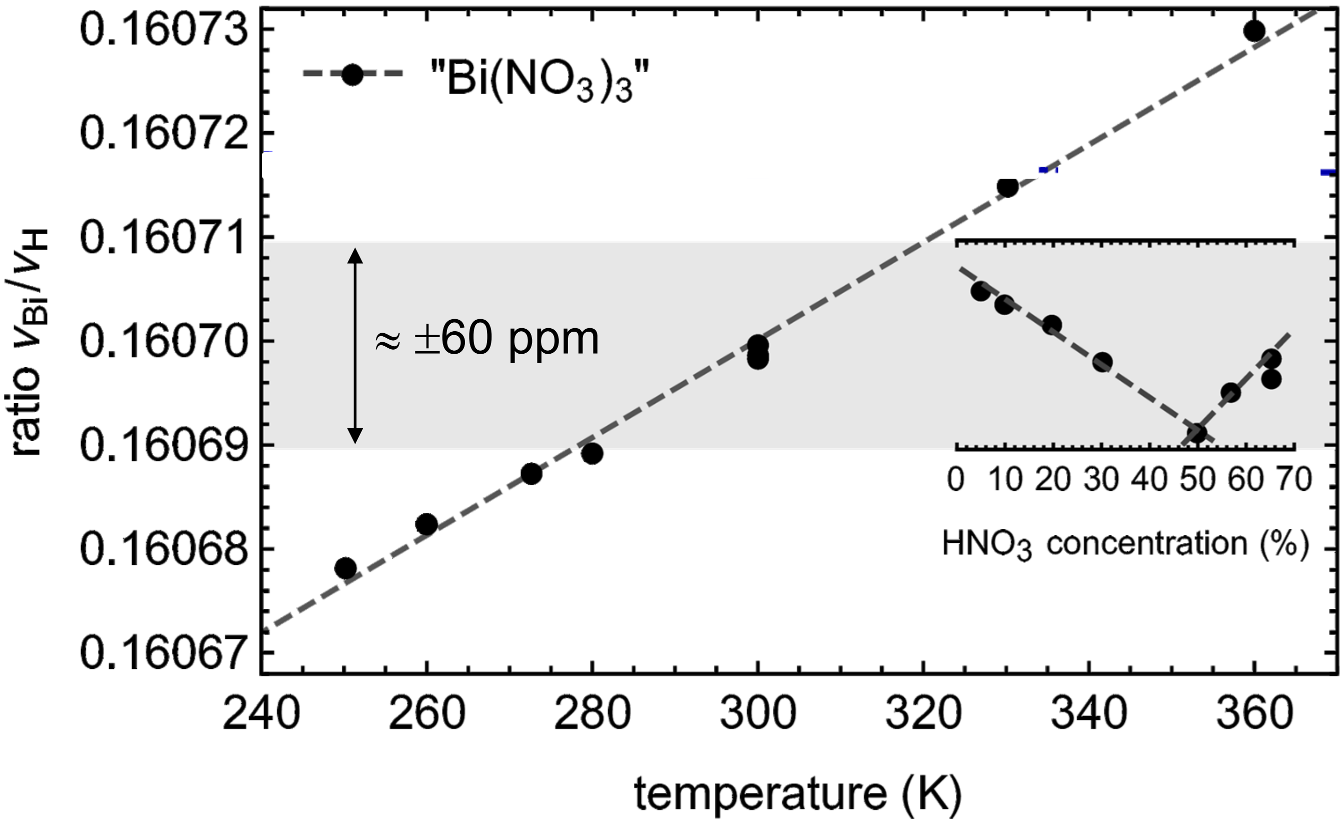
diamagnetic shielding



chemical shift



$[Bi(H_2O)_6]^{3+}$ cation

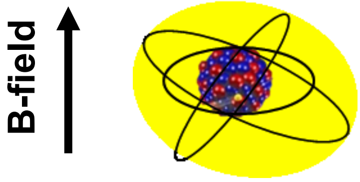


[Fedotov, M. A.; et al.
Neorg. Khim. 1998, 43, 307–310]

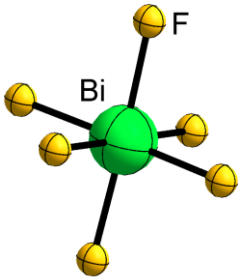
Chemical Shift in NMR Spectra of $Bi(NO_3)_3$ (aq)

$$\mu_{\text{Probe}} = \mu'_{\text{Probe}}/[1-(\sigma_{\text{dia}}+\sigma_{\text{chem}})]$$

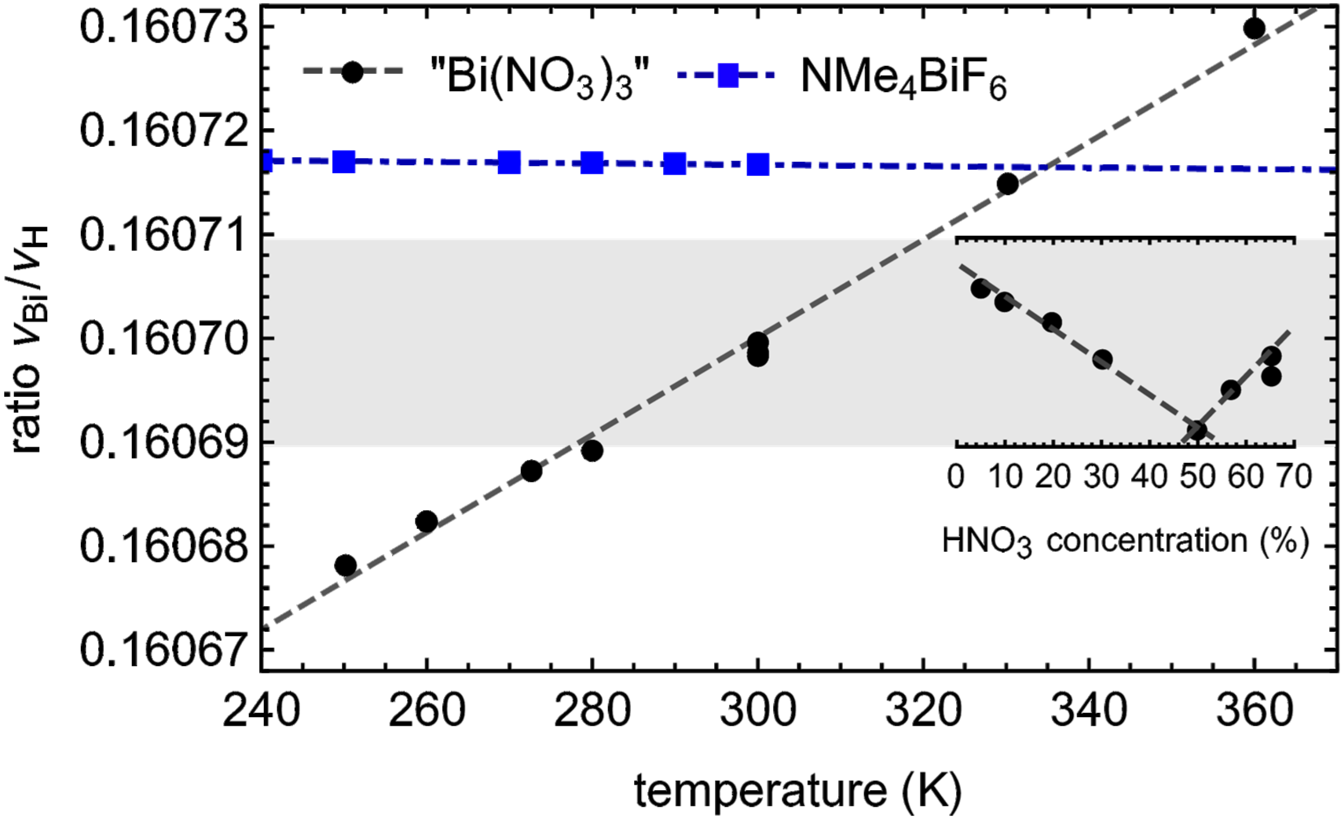
diamagnetic shielding



chemical shift



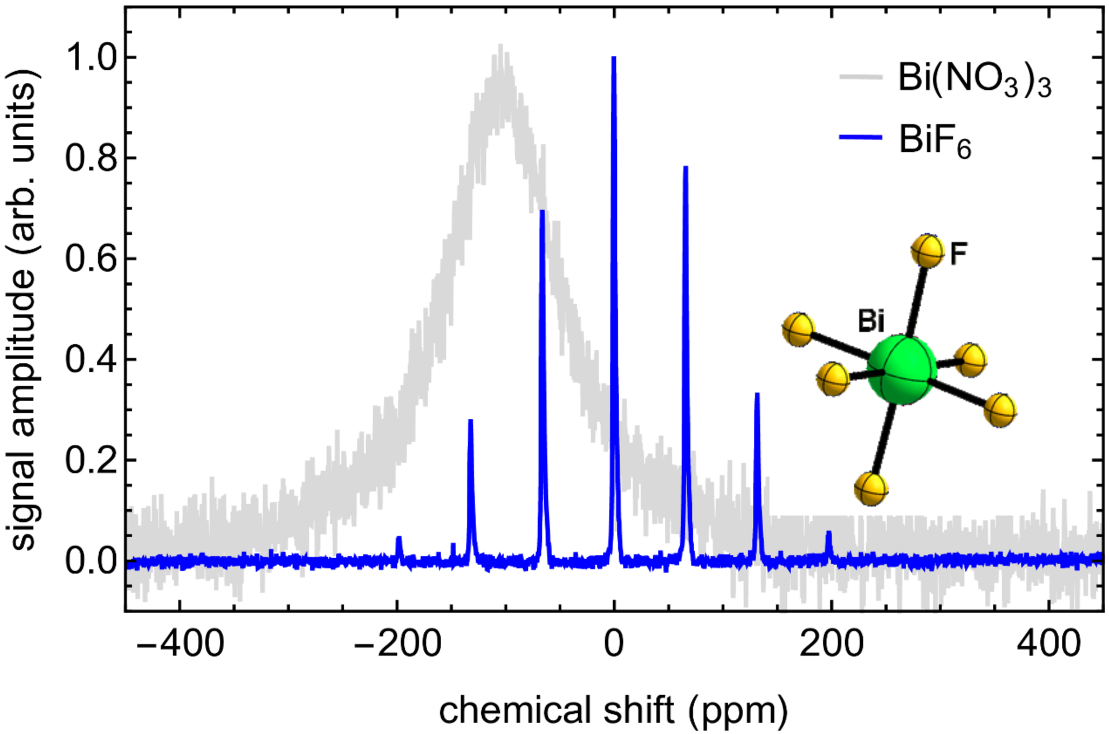
$[BiF_6]^-$ cation



[Fedotov, M. A.; et al.
Neorg. Khim. 1998, 43, 307–310]

Previous measurement of BiF_6^- : Morgan et al., J. Magn. Res. **52**, 139 (1969)

NMR Signals



$(\mathrm{CH_3})_4\mathrm{N}^+\mathrm{BiF}_6^-$ $(\mathrm{NMe_4BiF_6})$

Old:
$\mu(^{209}\mathrm{Bi}) = 4.1106(2)\ \mu_N$
Raghavan, At. Data Nucl. Data Tables **42**, 189 (1989)

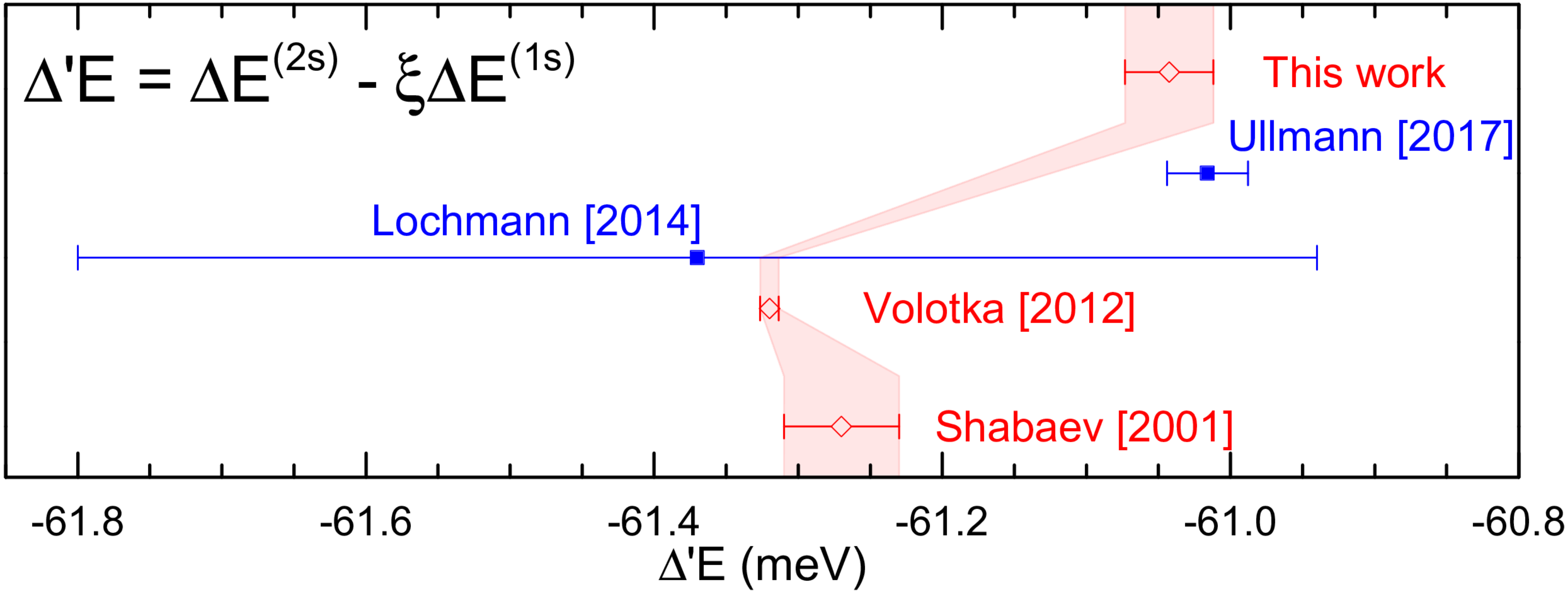
New:
$\mu(^{209}\mathrm{Bi}) = 4.092(2)\ \mu_N$
uncertainty dominated by theory

Diff. > $8\sigma_{\mathrm{comb}}$

	Theory		Experiment
	μ_I(old)	μ_I(new)	
$\Delta E^{(1s)}$	5112(-5/+20)	5089(-5/+20)(2)	5085.03(2)(9)
$\Delta E^{(2s)}$	801.9(-9/+34)	798.3(-9/+34)(4)	797.645(4)(14)

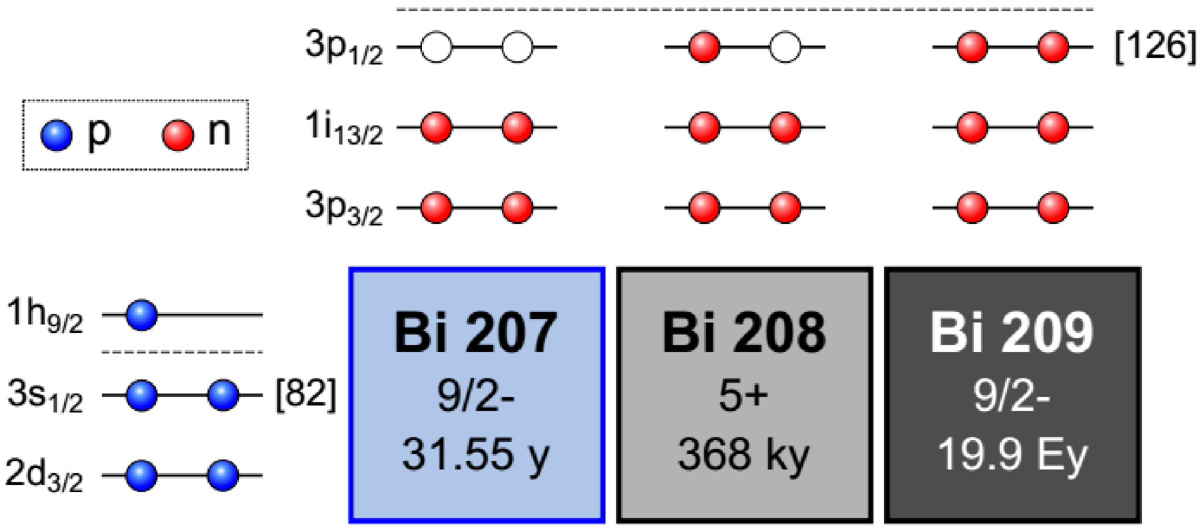
L. Skripnikov *et al.*, Phys. Rev. Lett. **120**, 093001 (2018)

Result for $\Delta' E$



L. Skripnikov *et al*., Phys. Rev. Lett. **120** 093001 (2018)

Bi Isotopes of Interest

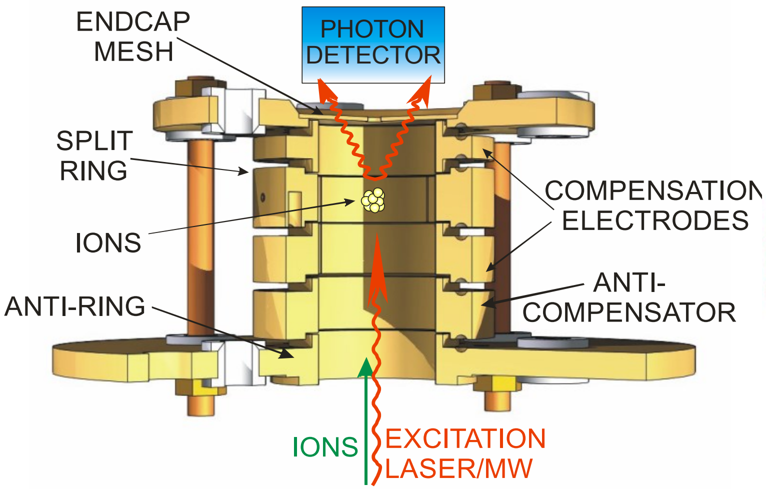


$\Delta E^{(1s)}$ (eV)	5.085	5.661	5.08503(9)
$\lambda^{(1s)}$ (nm)	244	**219**	243.8221(44)
$\Delta E^{(2s)}$ (eV)	0.7976	0.8894	0.797645(14)
$\lambda^{(2s)}$ (nm)	1554	**1394**	1554.377(29)

Towards Laser Spectroscopy of HCI in Ion Traps

- EBIT (MPIK)
  - Laser Fluorescence Spectroscopy (high T, pulsed lasers)
  - Mäckel *et al*., PRL **107**, 143002 (2011)
- SPECTRAP (GSI)
  - Fluorescence Spectroscopy at 4K, Doppler limited
  - Andelkovich *et al*., Phys. Rev. A **87**, 033423 (2013)
- ARTEMIS (GSI)
  - RF-Laser Fluorescence Double-Resonance Spectroscopy
  - Quint et al., Phys. Rev. A **78** 032517 (2008)
- αTRAP (MPIK)
  - Spin-flip Detection with Continuous Stern-Gerlach Effekt ($\approx$1K)
  - Egl *et al*., Phys. Rev. Lett. **123**, 123001 (2019)
- Cryogenic Paul Trap (PTB / MPIK)
  - Quantum Logic Spectroscopy (highest resolution)
  - Schmöger *et al*., Science **347**, 1233 (2015)
  - Micke *et al*., submitted to Nature (2019)

Long-Term Perspective at GSI

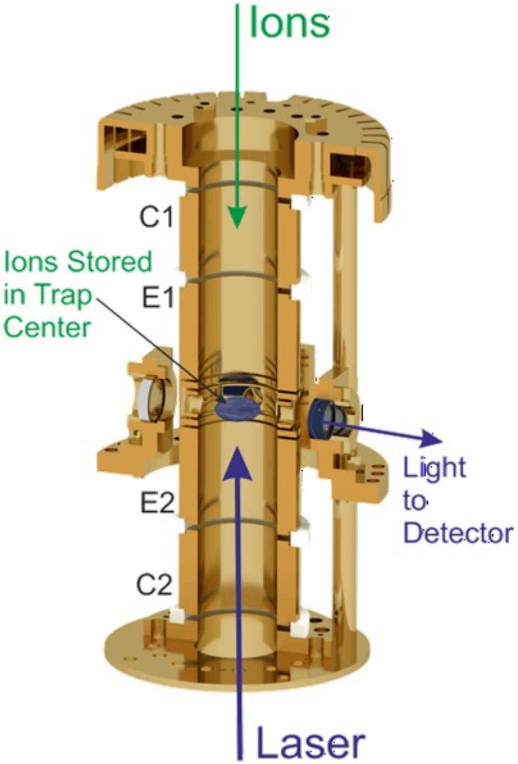


ARTEMIS

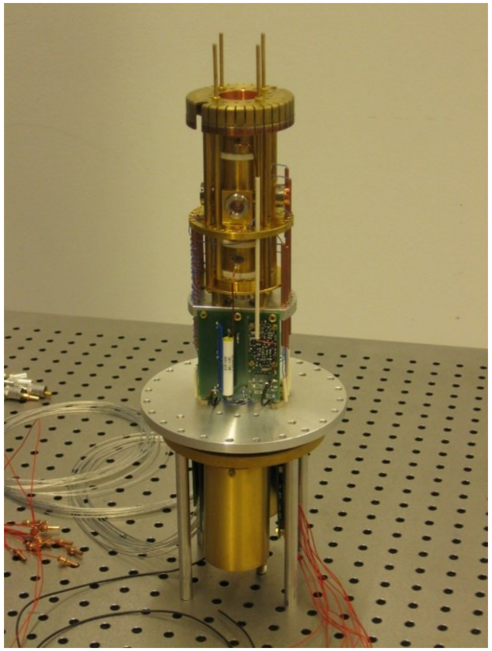
W. Quint, M. Vogel, G. Birkl, et al.

Technique:
Laser-microwave double resonance spectroscopy

- g-factor of bound electrons
- **nuclear g factor**
  (free of diamagnetic correction)
- atomic lifetimes
- higher-order Zeeman shifts



SPECTRAP

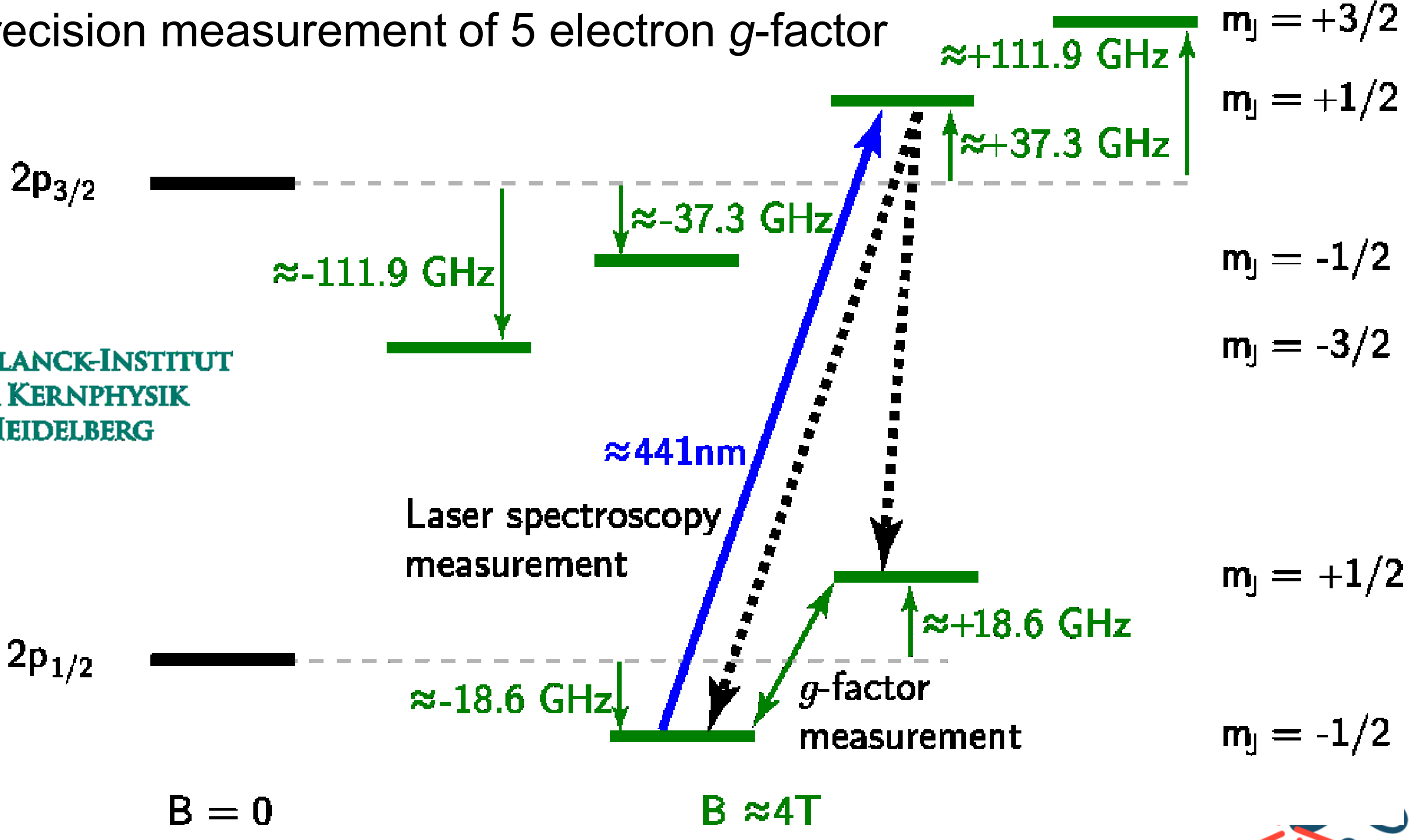


Z. Andelkovich, G. Birkl, M. Vogel, W. Nörtershäuser et al.

➔ Goal: Improved measurement of E(1s), E(2s) and $\Delta'E$
(gain: 2-3 orders of magnitude)

ALPHATRAP: Measurements on $^{40}\mathrm{Ar}^{13+}$

$^{40}\mathrm{Ar}^{13+}$ measurement of *g*-factor:
first high precision measurement of 5 electron *g*-factor

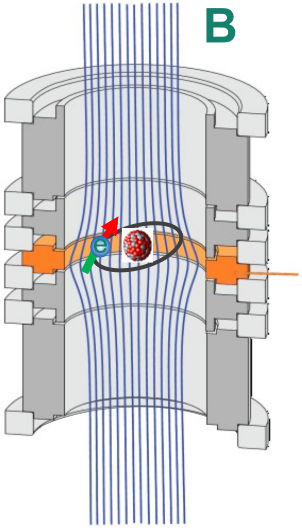


Courtesy of Alexander Egl

ALPHATRAP – Fluorescence-Free Detection

Continuous Stern-Gerlach Effect in a Penning Trap

Introducing magnetic bottle inhomogeneity



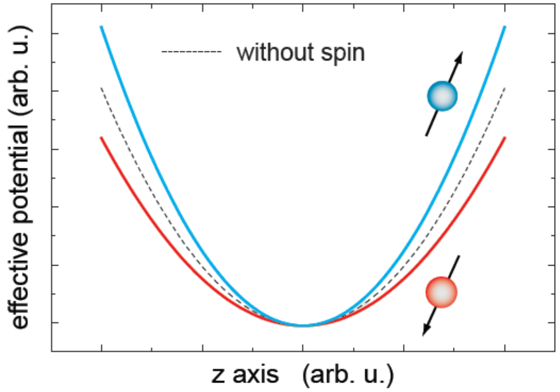
MAX-PLANCK-INSTITUT
FÜR KERNPHYSIK
HEIDELBERG

ferromagnetic
ring

Magnetic bottle
$B_2 \approx 45000$ T/m²

additional potential:

$$\Phi_z^{\mathrm{mag}} = \pm\mu_z\left[B_0 + B_2\left(z^2 - \frac{\rho^2}{2}\right)\right]$$



αTRAP

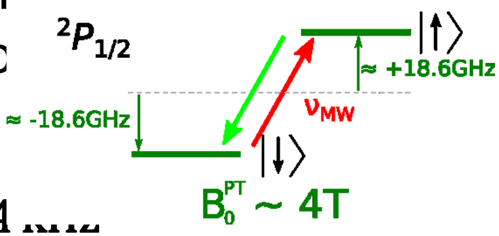
Courtesy of Alexander Egl

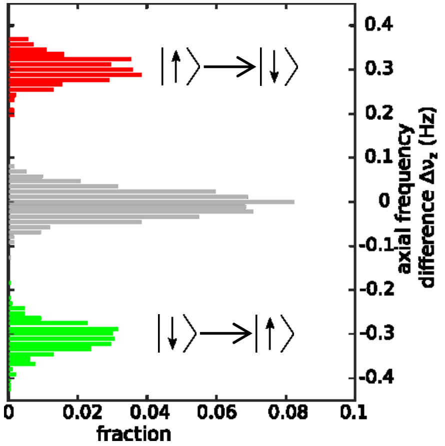
Continuous Stern-Gerlach effect:

axial frequency offset between
“up” and “down” spin orientatic

$$\boxed{\Delta\nu_z \approx \frac{B_2 g \mu_B}{4\pi^2 m_{ion}\nu_z}}$$

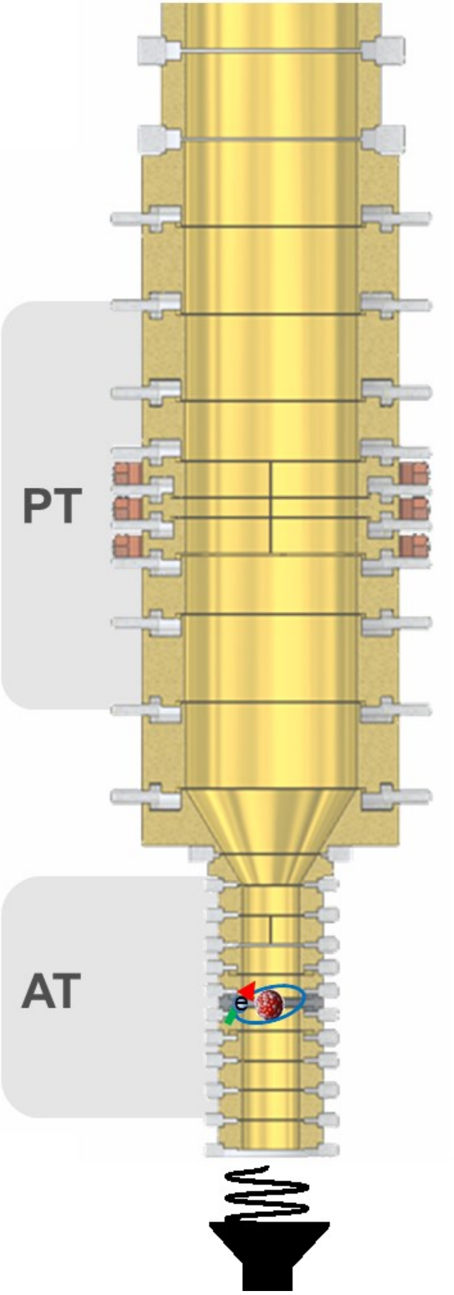
@ $\nu_z \approx 334$ kHz





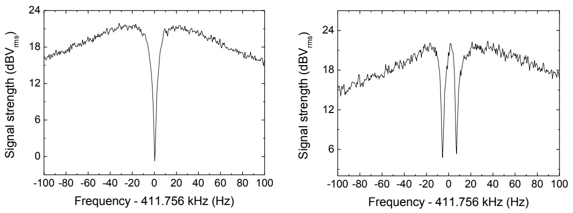
	$^{12}C^{5+}$	$^{28}Si^{13+}$	$^{40}Ar^{13+}$	$^{208}Pb^{81+}$
$\Delta\nu_z$	3.1 Hz	1.3 Hz	312 mHz	156 mHz

Measurement Cycle



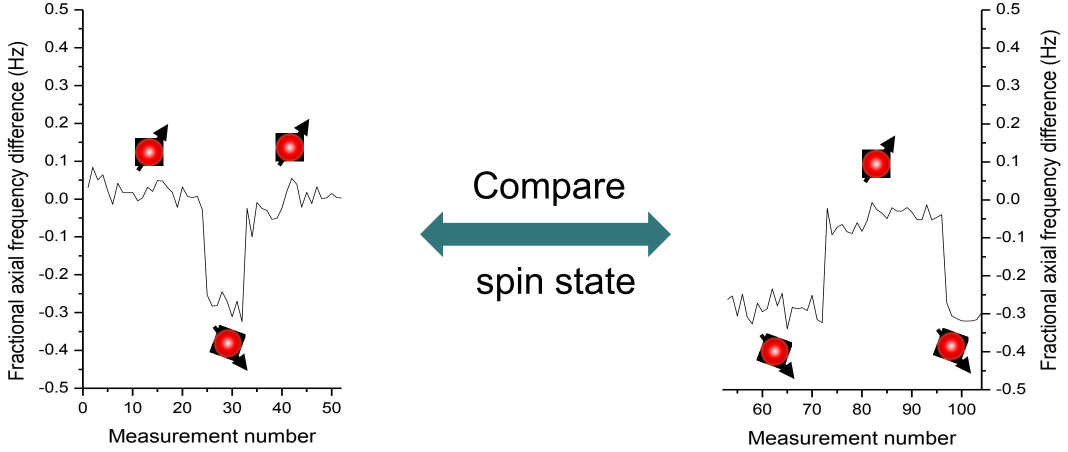
Courtesy of Alexander Egl

PT: Measurement of motional frequencies and probe with spectroscopy frequency



$$\nu_z^2 + \nu_+^2 + \nu_-^2 = \nu_c^2$$

AT: Detection and preparation of spin orientation



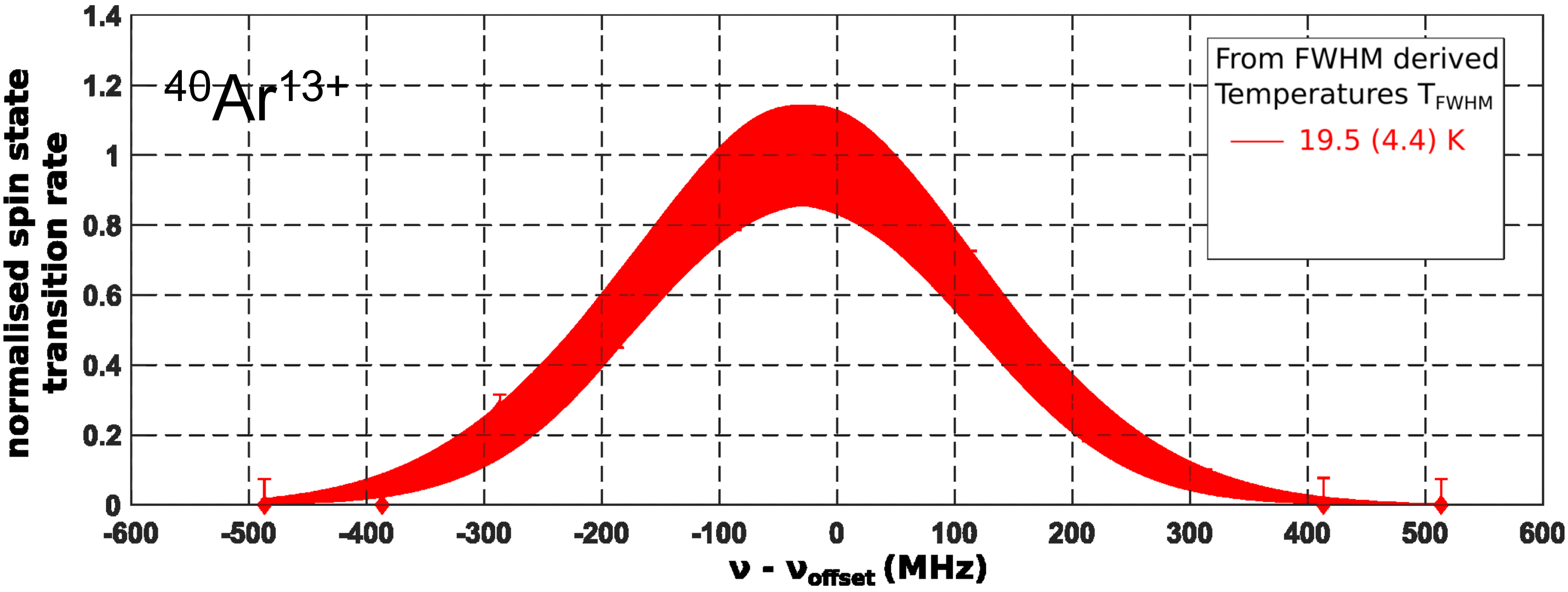
Compare

spin state

Laser Spectroscopy Applying Continuous Stern-Gerlach Effect

Results – Resonance for |1/2,-1/2> ↔ |3/2,+1/2> (= |J,mj>)
A. Egl, *et al*., PRL **123**, 123001 (2019)

- Derived temperature from FWHM → 19.5 ± 4.4 K



Courtesy of Alexander Egl

Laser Spectroscopy Applying Continuous Stern-Gerlach Effect

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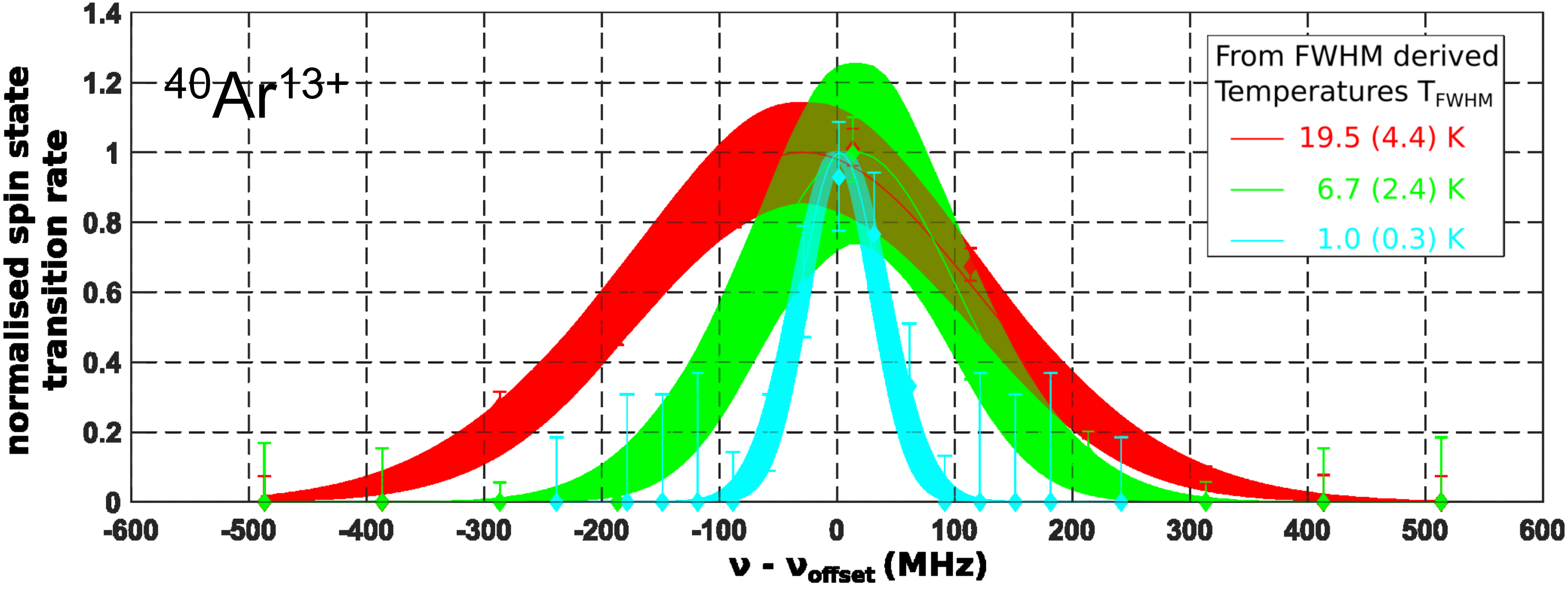
Results – Resonance for |1/2,-1/2> ↔ |3/2,+1/2> (= |J,mj>)
A. Egl, *et al.*, PRL **123**, 123001 (2019)

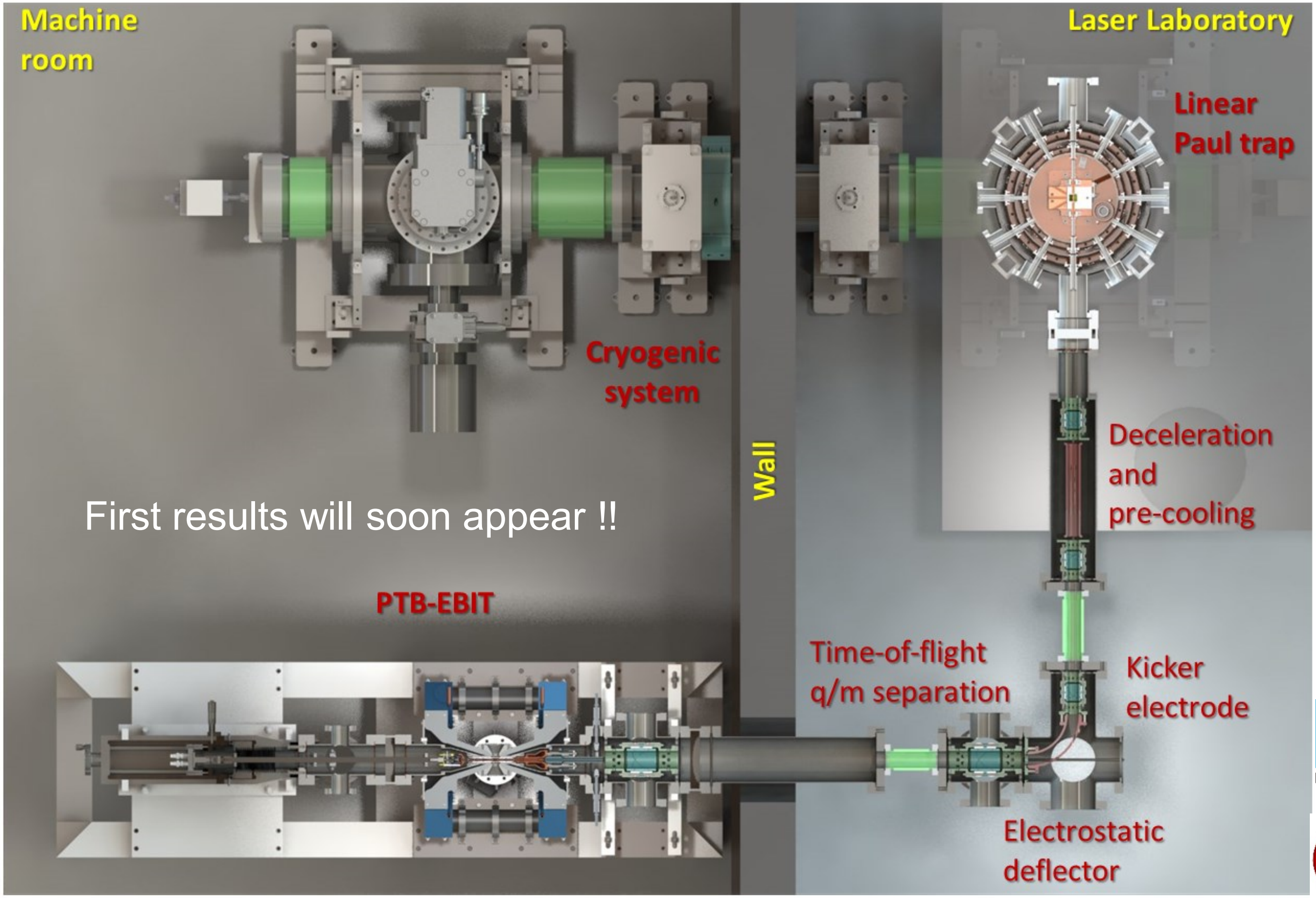
- Negative electronic feedback applied, expected lower temperature of factor 3
- Adiabatic cooling by lowering the trapping potential depth by a factor 3.8
  → lowered temperature by ≈ 3×3.8 = 11.4



ν = 679.216464 $(4)_{stat}(5)_{syst}$ THz, $\Delta\nu/\nu = 9\times10^{-9}$

Courtesy of Alexander Egl

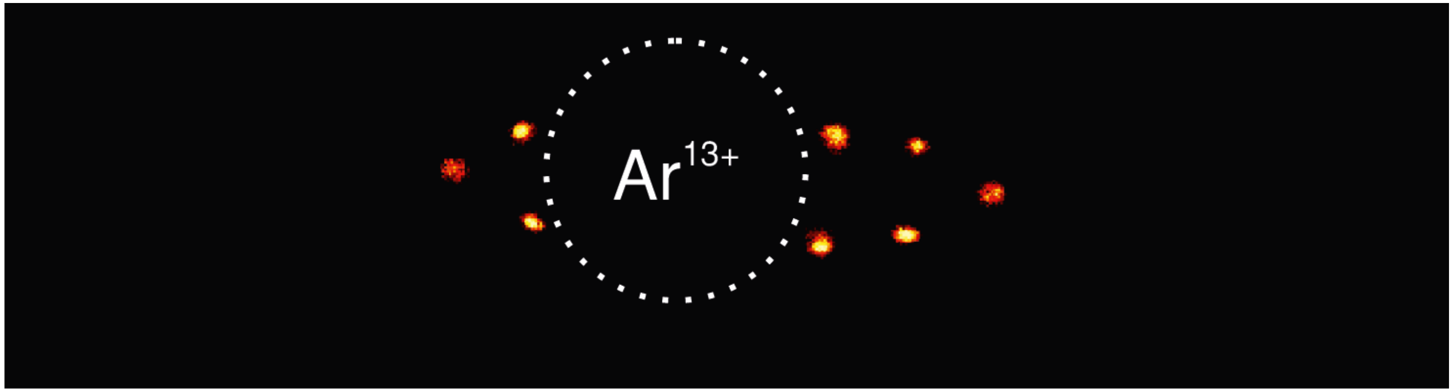
PTB-Experiment on cold HCI in a Paul-Trap

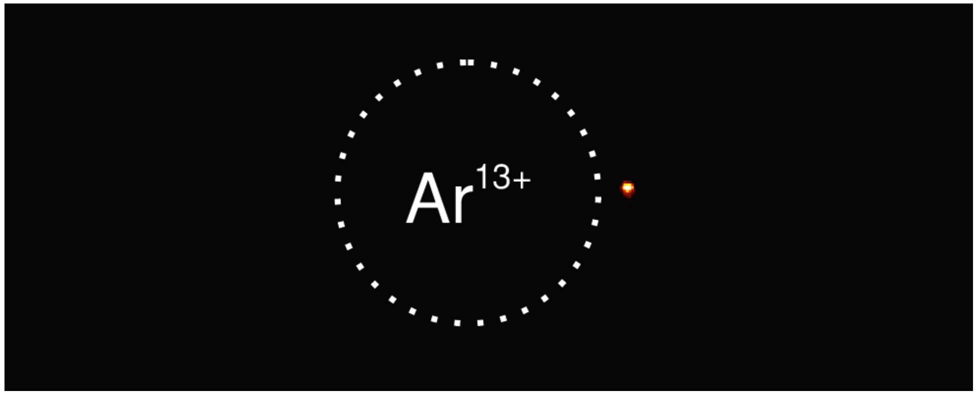


First results will soon appear !!

Courtesy of Peter Micke

Two-Ion Crystal Preparation





Schmöger et al., Science **347**, 1233 (2015)

Summary

- The measurement of $\Delta' E$ at the ESR strongly disagreed with QED calculations („HFS-Puzzle“).
- NMR Measurements and new chemical shift calculations in BiF_6^- were performed and proved that the Hyperfine Puzzle was caused by a wrong nuclear magnetic moment of ^{209}Bi.
- The measurements represents now a first (still inaccurate) test of bound-state QED in strong magnetic fields.

The road ahead of us:

- Determination of the nuclear magnetic moment of ^{208}Bi
- Prediction for the specific difference in ^{208}Bi
- Measurement of the hfs splitting in $^{208}Bi^{80+,82+}$ at the ESR storage ring → show indepedence of $\Delta' E$ from Bohr-Weisskopf effect
- TRAP measurements will provide higher accuracy (HITRAP → ARTEMIS & SPECTRAP, αTRAP, PTB-TRAP)

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HIC for FAIR
Helmholtz International Center

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