

Laser spectroscopy for nuclear structure physics

Ruben de Groote ruben.degroote@kuleuven.be Last lectures,

- We've discussed a little about the general principles of laser spectroscopy techniques
- You've seen examples of the physics we can study with magnetic moments, quadrupole moments and changes in mean-squared charge radii

This lecture,

- examples of the state-of-the-art in experimental techniques
  - In-source laser spectroscopy combined with Penning trap mass spectrometry the at IGISOL laboratory
  - Precision measurements: hyperfine structure beyond the quadrupole

Optical spectroscopy for nuclear structure research

#### Predominantly collinear fast-beam experiments



- Optical spectroscopy for nuclear structure research
- Pushing to lower production cross sections
  - in-source laser spectroscopy of silver

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  - in-source laser spectroscopy of silver
- When more precision is needed: collinear fastbeam laser spectroscopy
  - Laser spectroscopy of zinc

#### Predominantly collinear fast-beam experiments



**JYU.** Since 1863.

4.10.2021

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  - in-source laser spectroscopy of silver
- When more precision is needed: collinear fastbeam laser spectroscopy
  - Laser spectroscopy of zinc
- When even more precision is needed: beyond conventional optical spectroscopy
  - Future directions?

#### Predominantly collinear fast-beam experiments



JYU. Since 1863.

4.10.2021 6

- <sup>100</sup>Sn and the neighbouring isotopes are important isotopes to study in our quest to understand the atomic nucleus
- Producing these very neutron-poor isotopes is **challenging.**
- Reactions offer only small cross section, and other isotopes with the same mass (but closer to stability) are produced in much larger quantities.

	N=Z		50	Sn 99	Sn 100 1.16 s	Sn 101 1.97 s	Sn 102 3.8 s	Sn 103 7.0 s	Sn 104 20.8 s	Sn 105 34 s
	•	In 96	In 97 50 ms	In 98 37 ms	In 99 3.1 s	In 100 5.83 s	In 101 15.1 s	In 102 23.3 s	In 103 60 s	In 104 1.80 m
	Cd 94	Cd 95 90 ms	Cd 96 880 ms	Cd 97 1.10 s	Cd 98 9.2 s	Cd 99 16 s	Cd 100 49.1 s	Cd 101 1.36 m	Cd 102 5.5 m	Cd 103 7.3 m
Ag 92	Ag 93	Ag 94 37 ms	Ag 95 1.76 s	Ag 96 4.44 s	lg 97 5.5 s	Ag 98 47.5 s	Ag 99 2.07 m	Ag 100 2.01 m	Ag 101 11.1 m	Ag 102 12.9 m
Pd 91	Pd 92 1.1 s	Pd 93 1.15 s	Pd 94 9.0 s	Pd 95 7.5 s	Pd 96 122 s	Pd 97 3.10 m	Pd 98 17.7 m	Pd 99 21.4 m	Pd 100 3.63 d	Pd 101 8.47 h
		-			50	-				
	Projected reach of PI-ICR RIS				Optical measurement					

- I'd like to walk you through a recent experiment to show what it takes to perform measurements at the edges of our production capabilities
- Red: published work to date
- Gray: N=Z line, where nuclei have the same number of protons and neutrons

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	Projected reach of PI-ICR RIS				Optic meas	cal ureme	nt			

- I'd like to walk you through a recent experiment to show what it takes to perform measurements at the edges of our production capabilities
- Red: measured isotopes, some not yet published (look at how me have achieved over the past years!)
- Gray: N=Z line, where nuclei have the same number of protons and neutrons

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measurement



- Why does silver push out so much further than the other isotopes?
- At the heart of the answer: the atomic properties of silver.

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M. Reponen, R. P. de Groote et al, Nature Communications 12 (1), 1-8











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- Why does silver push out so much further than the other isotopes?
- At the heart of the answer: the atomic properties of silver
- Silver also doesn't stick very much inside of a thick target, which means the decay losses during extraction are minimal

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	Projected reach of PLICE RIS				Optical measurement					



- 1. Production
- <sup>14</sup>N(<sup>92</sup>Mo, 2p xn) Ag
- Ag ions stopped in graphite catcher foil
- Diffuse into hot cavity (few ms)





M. Reponen, R. P. de Groote et al, Nature Communications 12 (1), 1-8



#### M. Reponen, R. P. de Groote et al, Nature Communications 12 (1), 1-8



- 1. Production
- 2. Spectroscopy
- Laser ionization spectroscopy
- Possible to measure dipole moments and radii because the S<sub>1/2</sub> ground-state of silver possesses a very large A/µ-ratio



- 1. Production
- 2. Laser ionization spectroscopy

- 3. Detection and removal of contaminants
- Ions are injected into Penning trap
- Excitations of the ion motion in the trap
  - Kick out isobars
  - States in silver land on different areas of 2D-MCP detector



- 1. Production
- 2. Laser ionization spectroscopy
- 3. Detection
  - Count ions as function of laser frequency
  - Different gates on detector area select different nuclear state

#### Result:

Ultra clean background conditions

<sup>96</sup>Ag studied with an "on resonance" detection rate of **1 ion per 5 mins** 

Demonstrated for <sup>95</sup>Ag, too!



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Ultra clean background conditions

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Demonstrated for <sup>95</sup>Ag, too!

- Closest crossing of N=50 in this region to date
  - 2<sup>nd</sup> closest: Mo (*Z*=42)
- Event rate at the end of Penning trap: 0.005 events/s
- Background rate: ... 0?
- Remarkably sharp increase in radius observed
  - Comparison to state-of-the-art nuclear Fayans DFT is ongoing – but seems hard to explain with current theoretical tools



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# Predominantly collinear fast-beam experiments



#### Eye-watering higher orders.....



I.D. Moore, EJC 2021

#### PHYSICAL REVIEW A 103, 032826 (2021)

#### Magnetic octupole moment of <sup>173</sup>Yb using collinear laser spectroscopy

R. P. de Groote<sup>(0)</sup>,<sup>1,\*</sup> S. Kujanpää<sup>(0)</sup>,<sup>1</sup> Á. Koszorús<sup>(0)</sup>,<sup>2</sup> J. G. Li<sup>(0)</sup>,<sup>3</sup> and I. D. Moore<sup>(0)</sup>
<sup>1</sup>Department of Physics, University of Jyväskylä, PB 35(YFL) FIN-40351 Jyväskylä, Finland
<sup>2</sup>Department of Physics, University of Liverpool, Liverpool L69 7ZE, United Kingdom
<sup>3</sup>Institute of Applied Physics and Computational Mathematics, Beijing 100088, China



Defining K = F(F+1) - I(I+1) - J(J+1), this can be written as (truncated at the octupole (k = 3) term):  $E_F^{(1)} = \frac{AK}{2} + \frac{3B}{4} \frac{K(K+1) - I(I+1)J(J+1)}{(2I(2I-1)J(2J-1))} + \frac{5C}{4} \frac{K^3 + 4K^2 + \frac{4}{5}K(-3I(I+1)J(J+1) + I(I+1) + J(J+1) + 3) - 4I(I+1)J(J+1)}{I(I-1)(2I-1)J(J-1)(2J-1))},$ 

with hyperfine constants

$$A = \frac{1}{IJ} \langle II|T_2^{(n)}|II\rangle \langle JJ|T_1^{(e)}|JJ\rangle = \frac{\mu_I}{IJ} \langle JJ|T_1^{(e)}|JJ\rangle,$$
  

$$B = 4 \langle II|T_2^{(n)}|II\rangle \langle JJ|T_2^{(e)}|JJ\rangle = 2eQ\langle JJ|T_2^{(e)}|JJ\rangle,$$
  

$$C = \langle II|T_3^{(n)}|II\rangle \langle JJ|T_3^{(e)}|JJ\rangle = -\Omega \langle JJ|T_3^{(e)}|JJ\rangle.$$

$$H_{hyf} = \sum_{k} \mathbf{M}_{n}^{(k)} . \mathbf{T}_{e}^{(k)}$$

Extracting a nuclear moment requires knowledge of the atomic structure of the state in question!





Magnetic dipole

Magnetic octupole

Infinite expansion... Each term is significantly smaller than the previous one.



$$H_{hyf} = \sum_{k} \mathbf{M}_{n}^{(k)} \cdot \mathbf{T}_{e}^{(k)}$$
$$E_{F}^{(1)} = \sum_{k} (-1)^{I+J+F} \begin{cases} J & I & F \\ I & J & k \end{cases}$$
$$\times \langle I || M_{n}^{(k)} || I \rangle \langle J || T_{e}^{(k)} || J \rangle.$$





Magnetic dipole

Magnetic octupole



$$H_{hyf} = \sum_{k} \mathbf{M}_{n}^{(k)} \cdot \mathbf{T}_{e}^{(k)}$$

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$$\times \langle I || M_{n}^{(k)} || I \rangle \langle J || T_{e}^{(k)} || J \rangle.$$

$$E_{F}^{(1)} = \sum_{k} (-1)^{I+J+F} \frac{X_{i} \begin{cases} J & I & F \\ I & J & k \end{cases}}{\begin{pmatrix} I & I & F \\ I & J & k \end{pmatrix}}$$

Nuclear x atomic



...

$$A = \frac{\mu}{IJ} \times H(0)$$
$$B = 2eQ \times V_{zz}$$
$$C = -\Omega \times [?]$$
$$D = \Pi \times [??]$$

Ask your friendly neighbourhood atomic theorist what these are



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Unfortunate conventions...



Ask your friendly neighbourhood atomic theorist what these are



$$H_{hyf} = \sum_{\iota} \mathbf{M}_{n}^{(k)} \cdot \mathbf{T}_{e}^{(k)}$$
$$E_{F}^{(1)} = \sum_{k} (-1)^{I+J+F} \begin{cases} J & I & F \\ I & J & k \end{cases}$$
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$$E_F^{(1)} = \sum_k (-1)^{I+J+F} \frac{X_i \begin{cases} J & I & F \\ I & J & k \end{cases}}{\begin{pmatrix} I & 1 & I \\ -I & 0 & I \end{pmatrix} \begin{pmatrix} J & 1 & J \\ -J & 0 & J \end{pmatrix}}$$





$$\begin{split} H_{hyf} &= \sum_{\iota} \mathbf{M}_{n}^{(k)} \cdot \mathbf{T}_{e}^{(k)} \\ E_{F}^{(1)} &= \sum_{k} (-1)^{I+J+F} \begin{cases} J & I & F \\ I & J & k \end{cases} \\ &\times \langle I || M_{n}^{(k)} || I \rangle \langle J || T_{e}^{(k)} || J \rangle . \\ E_{F}^{(2)} &= \sum_{J'} \frac{1}{E_{J} - E_{J'}} \sum_{k_{1}, k_{2}} \begin{cases} F & J & I \\ k_{1} & I & J' \end{cases} \begin{cases} F & J & I \\ k_{2} & I & J' \\ k_{2} & I & J' \end{cases} \\ &\times \langle I || M_{n}^{(k_{1})} || I \rangle \langle I || M_{n}^{(k_{2})} || I \rangle \\ &\times \langle J' || T_{e}^{(k_{1})} || J \rangle \langle J' || T_{e}^{(k_{2})} || J \rangle , \end{split}$$

States with same F of different atomic levels mix





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These expressions may still look a little terrifying, but the stuff related to the angular momenta is actually the easy part.

The hard part lies in evaluating the atomic matrix elements!

# What is currently holding our experimental resolution/precision back?

+ **Fast** ( can be performed on short-lived isotopes ~ ms)

+ Sensitive (~ few ions/s)

+ Able to reach natural linewidth of strong transitions







#### Strong transition

-> Short half-life O(ns) -> Broad linewidth

Think back to the lecture on penning traps: short measurement time means low precision. It's really just fourier transforms.

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Think back to the lecture on penning traps: short measurement time means low precision. It's really just fourier transforms.

- In order to perform measurements with long interaction times, we need to store atoms or ions into a trap
- After previous lectures, you are all experts in how these work!
- Once in the trap, the ions can be carefully prepared
- Typically Paul traps, but penning traps are also possible.

36



# **Precision measure**

- In order to perform measureme interaction times, we need to st ions into a trap
- After previous lectures, you are how these work!
- Once in the trap, the ions can be carefully prepared
- Typically Paul traps, but penning traps are also possible.



- Barium has a simple atomic structure
- By using only two lasers, we can cool the ion down
  - Laser tuned slightly below resonance
  - photon is absorbed when the atom is moving towards the laser
  - Emission is isotropic: net effect means the ion slows down
  - Many cycles: reach temperatures of the order of Kelvin
  - Ask Franziska!





- Barium has a simple atomic structure
- By using only two lasers, we can cool the ion down
- Once the ion has been cooled, precise spectroscopy is possible
  - Radiofrequency excitations can be used to precisely measure splittings of the D states
  - Think of penning traps: long interaction, narrow resonance!







- The atom now sits very still, and can be gently examined with a laser beam
- Scanning over the optical transition...

<sup>40</sup> Optics Express Vol. 20, Issue 19, pp. 21379-21384 (2012)





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Table 6.2:  $5D_{5/2}$  hyperfine coupling constants.

- Combining the data with precise calculations by B. Sahoo
  - Relativistic coupled cluster (CCSD(T) method)

	A (Hz)	B(Hz)	C(Hz)
Uncorr.	-12029724.1(9)	59519566.2(43)	-41.73(18)
$\eta$ corr.	537(11)	5367(110)	_
$\zeta$ corr.	-46.9(12)	587(15)	29.33(75)
Corr.	-12029234(11)	59525520(110)	-12.41(77)

- Consistent value obtained from the two metastable D-states
- Nuclear theory interpretation of the result is yet to be made...

$$\Omega\left({}^{137}\mathrm{Ba}^{+}_{\mathrm{D}_{3/2}}\right) = 0.05057(54)~(\mu_{\mathrm{N}} \times \mathrm{b}),$$

$$\Omega\left({}^{137}\mathrm{Ba}^{+}_{\mathrm{D}_{5/2}}\right) = 0.0496(37)~(\mu_{\mathrm{N}}\times\mathrm{b}),$$

The fun doe	esn't even stop the	re		$\widehat{N} = \sum_{i=1}^{151} E u^{+} : {}^{9}S_{4} = M_{1}$
<ul> <li>External B-fasting!</li> <li>Direct us assign</li> </ul>	field: measure zee Inambiguous spin nent	man		H F=11/2 $\Delta m_r=0$ 11/2 $\Delta m_r=0$ 11/2 $\Delta m_r=0$ 11/2 -1/2 -1/2
hfs constant	<sup>151</sup> Eu <sup>+</sup> (Hz)	<sup>153</sup> Eu <sup>+</sup> (Hz)	<sup>151</sup> Eu <sup>+</sup> : <sup>153</sup> Eu <sup>+</sup>	
A B C D	$ \begin{array}{r} 1  540  297  394(13) \\ -  660  862(231) \\                                    $	$ \begin{array}{r}     684565993(9) \\     -1752868(84) \\     \hline     3(7) \\     -5(2) \end{array} $	2.250 034 927(35) 0.377 02(13) 9(22) 1.2(1.1)	0 100 200 300 400 500 rf Frequency + 10 017 200 (kHz)

Fit No.

VI

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# **Outline/conclusion**

- Optical spectroscopy for nuclear structure research
- Pushing to lower production cross sections
  - in-source laser spectroscopy of silver
- When more precision is needed: collinear fastbeam laser spectroscopy
  - Laser spectroscopy of zinc
- When even more precision is needed: beyond conventional optical spectroscopy
  - Future directions?
- Missing third axis: elements which are difficult to extract, and/or have complex atomic structure

# Predominantly collinear fast-beam experiments









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- Inject nuclei into an atom or ion trap
- Then, very long interaction times (seconds or more!) are possible
  - Drive forbidden optical transitions (linewidths << kHz)
  - Directly excite electrons within one atomic level (J, F, mF) → (J, F<sup>(·)</sup>, m<sub>F</sub><sup>(·)</sup>)

Measurement principle:

 Laser tuned to one transition will deplete an (F, m<sub>F</sub>) state after many cycles







- Laser tuned to one transition will deplete an (F, m<sub>F</sub>) state after many cycles
- Thus, fluorescence count rate will drop





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- Thus, fluorescence count rate will drop
- Radiofrequency field





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- Thus, fluorescence count rate will drop
- Radiofrequency field brings some electrons back!





- Laser tuned to one transition will deplete an (F, m<sub>F</sub>) state after many cycles
- Thus, fluorescence count rate will drop
- Radiofrequency field brings some electrons back!
- Fluorescence increases













Add external B-field: resolve Zeeman splitting Count the peaks, determine nuclear spin...

• Is it all worth the effort?

Fit No.	hfs constant	<sup>151</sup> Eu <sup>+</sup> (Hz)	<sup>153</sup> Eu <sup>+</sup> (Hz)	<sup>151</sup> Eu <sup>+</sup> : <sup>153</sup> Eu <sup>+</sup>
VI	A	1 540 297 394(13)	684 565 993(9)	2.250 034 927(35)
	B	-660862(231)	-1752868(84)	0.377 02(13)
	С	26(23)	3(7)	9(22)
	D	-6(5)	-5(2)	1.2(1.1)

