

Shining light on neptunium

Laser spectroscopy for probing atomic structure

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Neptunium

- Radioactive actinide
- Long half-life ²³⁷Np 2.14 · 10⁶ y
- High radiotoxicity



S. Kohler et al. Spectrochim. Acta B 52, 717 – 726, (1997)
 V. Kazakov et al. Phys. Scr. 92, 10, (2017)
 N.J. Stone, At. Data Nucl. Data Tables 90, 1, 75-176, (2005)

- Ionization potential 50 535(2) cm^{-1 [1]}
 6.26554(25) eV
- 462 atomic levels known ^[2]
- ²³⁷Np
 - Magnetic dipole moment +3.14(4) $\mu_{N}^{[3]}$
 - Electric quadrupole moment +3.886(4) *e*b ^[3]
- ²³⁹Np
 - Nuclear moments unknown



Neptunium production and trace analysis







Trace analysis of environmental samples is of high relevance

- ²³⁹Np as a tracer for precise quantification
- Identification of efficient ionization schemes:
 - High elemental selectivity
 - Isotope related effects like hyperfine structures (HFS) and isotope shift (IS)

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large splitting and shifts observed in neptunium





Neptunium - on-line studies

- On-line studies of neptunium at ISOLDE
 - In-source laser resonance ionization spectroscopy of neptunium and plutonium (*Letter of Intent INTC-I-243*)
- Access to other isotopes
- Nuclear moments are only known for ²³⁷Np
- Validation of theoretical estimates

Need for efficient and selective ionization schemes









Technique and setup

Resonance ionization spectroscopy The RISIKO mass separator Laser ion source





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Probing atomic structure with lasers



- Atomic level structure through optical transitions
- Ionization limit through Rydberg convergences
- Nuclear spins and electromagnetic moments through hyperfine interaction





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Resonance ionization mass spectrometry





RISIKO mass separator





8



RISIKO laser ion source

- High operation temp. up to about 2000 °C
- Contamination caused by:
 - black body radiation
 - collisions
- Doppler broadening limiting spectral resolution

$$\Delta v = v_0 \sqrt{\frac{8k_B T \ln 2}{mc^2}} \sim \text{GHz}$$

 Δv - linewidth k_B - Boltzmann constant

 m^- atom mass

 v_0 - resonance freq. for a particle at rest

- T temperature
- c speed of light



Sample







Probing atomic structure

Scheme development Line profiles Ionization potential determination Development and characterization of FI-LIST

Np 237 2.14·10⁶ a α 4.790...;

29...; e-



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[1] M. Kaja et al., Eur. Phys. J. D 78, 50 (2024)



[1] M. Kaja et al., Eur. Phys. J. D, 78, 50 (2024)

12









Spectra below the ionization potential

[1] M. Kaja et al., Eur. Phys. J. D, 78, 50 (2024)

14



Assignment of levels or analysis of Rydberg Convergences fails for such a complex atomic systems



Field ionization - saddle point model



 $Z_{\rm eff}$ - effective charge *F* - external field strength ε_0 - vacuum permittivity

• Extrapolation to F = 0 V/cm yields IP





- Well controllable electric field for ionization between el2 and el3
- Missing energy to ionize an atom is from the electric field F = (el2 el3)/1 cm



Characterization of FI-LIST – ytterbium measurements



H. Lehec et al., Phys. Rev. A **98**, 062506, (2018)
 R. Li et al., Spectrochim. Acta B 174 (2020)
 M. Kaja et al., NIM B **547**, 165213 (2024)



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Ytterbium Ionization Potential



Ytterbium Ionization Potential



Ionization potential

[1] M. Kaja et al., Eur. Phys. J. D 78, 50 (2024)







Ionization potential



[1]S. Köhler et al., Spectrochim. Acta B 52 (6) (1997)
[2] M. Kaja et al., Eur. Phys. J. D 78, 50 (2024)

21



Ionization potential



Np 239

2.355 d

γ 106...; e⁻; g

β-**0.4**...

High-resolution spectroscopy

Hyperfine structure measurements Extraction of nuclear moments

JGU

 Np 237

 2.14·10⁶ a

 α 4.790...;

 γ 29...; e⁻

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

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PI-LIST - Perpendicularly Illuminated LIST

- **Separation** of hot cavity evaporation from laser ionization volume
- **Suppression** of surface ionized species
- **Pulsed laser ionzation** inside RF quadrupole structure
- **Reduced Doppler width** in laser transversal interaction

[1] R. Heinke et al., Hyperfine Interact 238, 6 (2017)
[2] R. Heinke, ..., M. Kaja et. al., NIM B, 541, 8-12, (2023)

24

[1] M. Kaja et al., Eur. Phys. J. D 78, 50 (2024)

Hyperfine spectroscopy in ^{237}Np (I = 5/2)

Neptunium production and trace analysis

neutron

capture

β⁻ decay

Trace analysis of environmental samples is of high relevance

- ²³⁹Np as a tracer for precise quantification
- Identification of efficient ionization schemes:
 - High elemental selectivity
 - Isotope related effects like hyperfine structures (HFS) and isotope shift (IS)
 - large splitting and shifts observed in neptunium

Sample:

- 10¹³ atoms of ²³⁷Np
- 10¹¹ atoms of ²³⁹Np

²³⁹Np production:

- irradiation of ²³⁸U at
- reactor TRIGA Mark II Mainz

²³⁷Np – stock solution

Hyperfine spectroscopy in ²³⁷Np and ²³⁹Np

Hyperfine parameters and nuclear moments

Conclusion

- Atomic structure investigations of neptunium
 - Ionization schemes, atomic energy levels, lifetimes
 - Precise IP determination of 50 535.54(15) cm⁻¹
 - *M. Kaja et al., Eur. Phys. J. D* **78**, 50 (2024)
- The FI-LIST a new useful type of the PI-LIST
 - Dedicated tool for IP studies on rare elements with complex structure
 - Suitable off-line as well as on-line applications
 - Capable of measurements at very low electric fields of ~ 1 V/cm
 - *M. Kaja et al., NIM B* **547**, 165213 (2024)
- High resolution spectroscopy in ²³⁷Np and ²³⁹Np
 - Isotope shifts
 - Nuclear moments for ²³⁹Np
 - *M. Kaja et al., Eur. Phys. J.* A **60**, 140 (2024)

Outlook

- Off- and on-line IP determination of other elements with complex spectra, e.g. Fm
- Extension of the high-resolution spectroscopy to other isotopes of neptunium

Thank you!

LISA conference 02.09.2024 - Magdalena Kaja

Technique: Resonance ionization mass spectrometry

Lifetime investigations of first excited states

[1] M. Kaja et al., Eur. Phys. J. D 78, 50 (2024)

The excited-state population decay as a function of the ionization-pulse delay.

Population development in the "dark" time between pulses corresponds to an exponential distribution.

This method is applicable to lifetimes much longer than the laser-pulse duration (\approx 50ns) and much shorter than the collision lifetime of the excited atoms within the laser beam in the atomizer tube(\approx 3µs)

Ytterbium

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Laser Ion Source and Trap (LIST)

Perpendicularly Illuminated LIST **PI-LIST**

Field Ionization LIST FI-LIST

Ti:Sapphire lasers system

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LISA conference 02.09.2024 - Magdalena Kaja

LIS/

Neptunium

- Radioactive actinide
- Long half-life ²³⁷Np 2.14 · 10⁶ y
- High radiotoxicity

Neptunium production

Kohler, S; et al. Spectrochim. Acta B,52, 717 – 726, (1997)
 Kazakov, V; et al. Phys. Scr., 92, 10, (2017)
 Stone, N.J. At. Data Nucl. Data Tables, 90, 1, 75-176, (2005)

The development of efficient and selective laser ionization schemes plays an important role for Np spectroscopy and trace analysis.

It is important to take into account the isotope-related effects in ionization schemes coming from hyperfine structure (HFS) and isotope shift (IS).

²³⁹Np

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• Moments unknown

LARISA LAB

Laser development system 3

including seeding and direct laserdiode pumping

RISIKO –

High voltage sector field MS for high efficiency, high sensitivity spectroscopy and ion implantation

> Ti:Sa Laser system 2

Ti:Sa Laser system 1

MABU – low voltage quadrupole MS for laser spectroscopy

RISIKO mass separator

LISA conference 02.09.2024 - Magdalena Kaja

Courtesy of Reinhard Heinke and Asar AH Jaradat

Robust metallic mirrors

"Sub-Doppler" Hot Cavity In-source Spectroscopy

- Crossed atom beam / laser geometry in LIST structure
- Selection of **reduced Doppler ensemble** in laser intersection volume

Adapted from R. Heinke PhD thesis

• Suitable narrow-band laser

Courtesy of Reinhard Heinke and Asar AH Jaradat

Actinium high resolution sprectroscopy

Courtesy of Reinhard Heinke and Asar AH Jaradat

Probing atomic structure with lasers

- Atomic level structure through optical transitions
- Ionization limit through Rydberg convergences
- Nuclear spins and electromagnetic moments through hyperfine interaction

n – principal quantum number S L – total orbital angular momentum I

Q – electric quadrupole moment

J – total angular momentum

- S total spin angular momentum n I – nuclear spin
- μ magnetic dipole moment

44

Kohler, S; et al. Spectrochim. Acta B,52, 717 – 726, (1997)
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 Stone, N.J. At. Data Nucl. Data Tables, 90, 1, 75-176, (2005)

FI-LIST Field Ionization LIST

HFS result and nuclear moments in Np

		Fit		Literature [1]			μ_{I} [μ_{N}]	Q_s [barn]		
p 237	FES [cm ⁻¹]	A [MHz]	B [MHz]	A [MHz]	B [MHz]	J	+3.14(4) [2]	+3.886(4) [2]		
.14 [.] 10 ⁶ a	0	776.10(18)	929(5)	778(10)	645(100)	11/2	[2] Stone, N.J. At. Data Nucl. Data Tables, 90, 1, 75-176, (2005)			
4.790; !9; e⁻	25 075.1	1470.02(10)	327(5)	1470(10)	264(100)	13/2				
	25 277.64	570.08(14)	-307(4)	X	X	9/2				

		F		
Nn 239	FES [cm ⁻¹]	A [MHz]	B [MHz]	J
2 355 d	0	785.26(39)	949(10)	11/2
β ⁻ 0.4	25 075.15	1487.06(25)	336(9)	13/2
y 100, e , g	25 277.63	576.74(40)	-319(11)	9/2

Determination of nuclear moments

[1] Fred. M: et al. J. Opt. Soc. Am., 67, 1 (1977)

Presented results are preliminary, to be published by M. Kaja et al.

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Larissa activities on laser spectroscopy in the actinides

Ionization potential determination of curium

High resolution spectroscopy of californium

F. Weber,.., M.K., et al., Phys. Rev. C 107, 034313 (2023)

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