

The heaviest ever heavy element? Lawrencium

LISA Conference 2024 Geneva

J. S. Andrews¹ J. Grumer² S. Fritzsche¹ A. Bondaraev¹
P. Jönsson³ J. Bieroń⁴

¹Helmholtz-Institut Jena, Germany ²Uppsala Universitet, Sweden

³Malmö Universitet, Sweden ⁴Uniwersytet Jagielloński, Poland

2 September 2024

HI JENA
HELMHOLTZ
Helmholtz-Institut Jena

Table of Contents

- ① Introduction
- ② Experiment
- ③ Theory

- ④ Methods
- ⑤ Results
- ⑥ Conclusion



Heaviest ever heavy element?

1 H Hydrogen Nonmetal																	2 He Helium Noble Gas						
3 Li Lithium Alkali Metal	4 Be Beryllium Alkali Earth Metal																	5 B Boron Metalloid	6 C Carbon Nonmetal	7 N Nitrogen Nonmetal	8 O Oxygen Nonmetal	9 F Fluorine Halogens	10 Ne Neon Noble Gas
11 Na Sodium Alkali Metal	12 Mg Magnesium Alkali Earth Metal																	13 Al Aluminum Post-transition Metal	14 Si Silicon Metalloid	15 P Phosphorus Nonmetal	16 S Sulfur Nonmetal	17 Cl Chlorine Halogens	18 Ar Argon Noble Gas
19 K Potassium Alkali Metal	20 Ca Calcium Alkali Earth Metal	21 Sc Scandium Transition Metal	22 Ti Titanium Transition Metal	23 V Vanadium Transition Metal	24 Cr Chromium Transition Metal	25 Mn Manganese Transition Metal	26 Fe Iron Transition Metal	27 Co Cobalt Transition Metal	28 Ni Nickel Transition Metal	29 Cu Copper Transition Metal	30 Zn Zinc Transition Metal	31 Ga Gallium Post-transition Metal	32 Ge Germanium Metalloid	33 As Arsenic Metalloid	34 Se Selenium Nonmetal	35 Br Bromine Halogens	36 Kr Krypton Noble Gas						
37 Rb Rubidium Alkali Metal	38 Sr Strontium Alkali Earth Metal	39 Y Yttrium Transition Metal	40 Zr Zirconium Transition Metal	41 Nb Niobium Transition Metal	42 Mo Molybdenum Transition Metal	43 Tc Technetium Transition Metal	44 Ru Ruthenium Transition Metal	45 Rh Rhodium Transition Metal	46 Pd Palladium Transition Metal	47 Ag Silver Transition Metal	48 Cd Cadmium Transition Metal	49 In Indium Post-transition Metal	50 Sn Tin Post-transition Metal	51 Sb Antimony Metalloid	52 Te Tellurium Metalloid	53 I Iodine Halogens	54 Xe Xenon Noble Gas						
55 Cs Cesium Alkali Metal	56 Ba Barium Alkali Earth Metal		72 Hf Hafnium Transition Metal	73 Ta Tantalum Transition Metal	74 W Tungsten Transition Metal	75 Re Rhenium Transition Metal	76 Os Osmium Transition Metal	77 Ir Iridium Transition Metal	78 Pt Platinum Transition Metal	79 Au Gold Transition Metal	80 Hg Mercury Transition Metal	81 Tl Thallium Post-transition Metal	82 Pb Lead Post-transition Metal	83 Bi Bismuth Metalloid	84 Po Polonium Metalloid	85 At Astatine Halogens	86 Rn Radon Noble Gas						
87 Fr Francium Alkali Metal	88 Ra Radium Alkali Earth Metal		104 Rf Rutherfordium Transition Metal	105 Db Dubnium Transition Metal	106 Sg Seaborgium Transition Metal	107 Bh Bohrium Transition Metal	108 Hs Hassium Transition Metal	109 Mt Meitnerium Transition Metal	110 Ds Darmstadtium Transition Metal	111 Rg Roentgenium Transition Metal	112 Cn Copernicium Transition Metal	113 Nh Nihonium Post-transition Metal	114 Fl Flerovium Post-transition Metal	115 Mc Moscovium Post-transition Metal	116 Lv Livermorium Post-transition Metal	117 Ts Tennessine Halogens	118 Og Oganesson Noble Gas						
		57 La Lanthanum Lanthanide	58 Ce Cerium Lanthanide	59 Pr Praseodymium Lanthanide	60 Nd Neodymium Lanthanide	61 Pm Promethium Lanthanide	62 Sm Samarium Lanthanide	63 Eu Europium Lanthanide	64 Gd Gadolinium Lanthanide	65 Tb Terbium Lanthanide	66 Dy Dysprosium Lanthanide	67 Ho Holmium Lanthanide	68 Er Erbium Lanthanide	69 Tm Thulium Lanthanide	70 Yb Ytterbium Lanthanide	71 Lu Lutetium Lanthanide							
		89 Ac Actinium Actinide	90 Th Thorium Actinide	91 Pa Protactinium Actinide	92 U Uranium Actinide	93 Np Neptunium Actinide	94 Pu Plutonium Actinide	95 Am Americium Actinide	96 Cm Curium Actinide	97 Bk Berkelium Actinide	98 Cf Californium Actinide	99 Es Einsteinium Actinide	100 Fm Fermium Actinide	101 Md Mendelevium Actinide	102 No Nobelium Actinide	103 Lr Lawrencium Actinide							

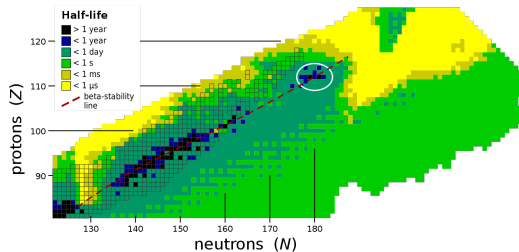
Superheavy elements

Heavy elements



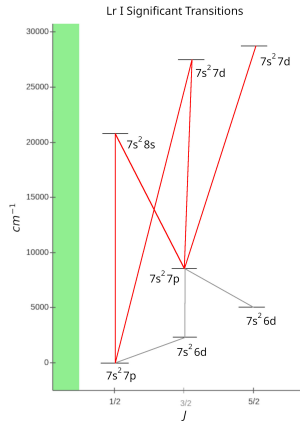
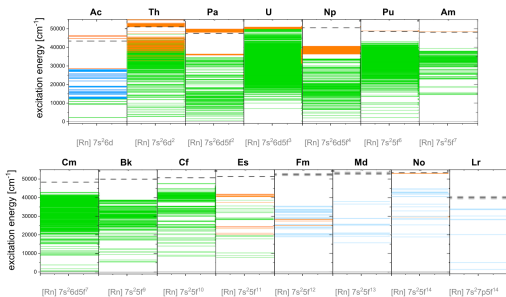
Island of stability

- We are interested in searching for the island of stability, defining new frontiers in nuclear physics¹
- This can be done by testing nuclear models by investigating the hyperfine structure
- But... we need to confirm the broad structure first!



¹M. Block et al., Progress in Particle and Nuclear Physics **116**, 103834 (2021).

Lr and the actinides



Applications

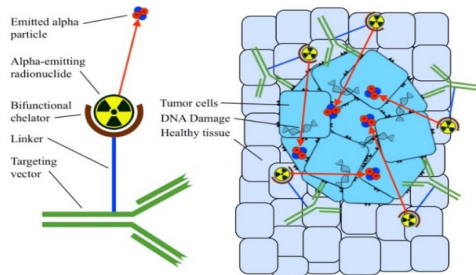
The main scientific motivation to study Lr is driven by an interest in the study of nuclear, relativistic effects and electron correlations. These strongly influence the atomic structure.

²B. J. B. Nelson et al., *Pharmaceutics* **13**, 49 (2020).

Applications

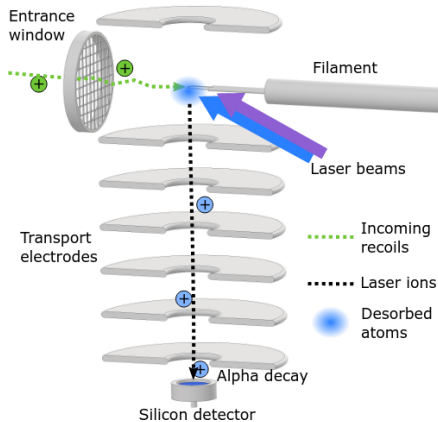
The main scientific motivation to study Lr is driven by an interest in the study of nuclear, relativistic effects and electron correlations. These strongly influence the atomic structure.

- Nuclear medicine is a rapidly emerging field.²
- Targeted alpha therapy (TAT)
- α -emitting radionuclides attached to targeting vectors to treat various diseases.



²B. J. B. Nelson et al., *Pharmaceutics* **13**, 49 (2020).

Experimental setup



³J. Warbinek et al., *Atoms* **10**, 41 (2022).

Hartree-Fock

The energy levels can be obtained by solving the eigenvalue problem

$$\hat{H}\Psi = E\Psi \quad (1)$$

The HF wavefunction is modelled as a single Slater determinant

Atom	Obs.	HF	Diff.
<i>n</i> = 2 shell			
Li	5.39	5.34	0.05
Be	9.32	8.42	0.90
B	8.30	8.43	-0.13
C	11.26	11.79	-0.53
N	14.53	15.44	-0.91
O	13.62	14.45	-0.85
F	17.42	18.62	-1.20
Ne	21.56	23.14	-1.58

Observed and HF ionisation potentials
for neutral atoms [eV]

Hartree-Fock

The energy levels can be obtained by solving the eigenvalue problem

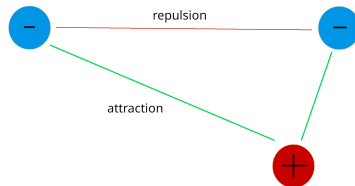
$$\hat{H}\Psi = E\Psi \quad (1)$$

The HF wavefunction is modelled as a single Slater determinant

Atom	Obs.	HF	Diff.
<i>n</i> = 2 shell			
Li	5.39	5.34	0.05
Be	9.32	8.42	0.90
B	8.30	8.43	-0.13
C	11.26	11.79	-0.53
N	14.53	15.44	-0.91
O	13.62	14.45	-0.85
F	17.42	18.62	-1.20
Ne	21.56	23.14	-1.58

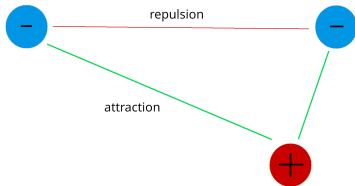
Observed and HF ionisation potentials
for neutral atoms [eV]

- HF usually does not agree with experiment



- HF is a mean-field theory and does not fully consider electron-electron interactions

MCDHF



Electron correlation is defined by

$$E_{corr} = E_{exact} - E_{HF} \quad (2)$$

To account for electron correlation, additional electronic configurations are included. In multi-configurational Dirac-Hartree Fock (MCDHF), a wavefunction is created by a linear combination of

configuration state functions (CSFs)

$$\Psi(\gamma\pi JM) = \sum_{i=1}^{N_{CSFs}} c_i \psi_i(\gamma_i\pi JM) \quad (3)$$

where π , J , M , γ_i is the parity, total angular momentum and magnetic quantum number. γ_i describes other numbers needed to uniquely describe the CSF.

- A CSF is a symmetry adapted linear combination of Slater determinants
- The Dirac-Colomb Hamiltonian is used.
- Breit interaction and further QED effects are added perturbationally

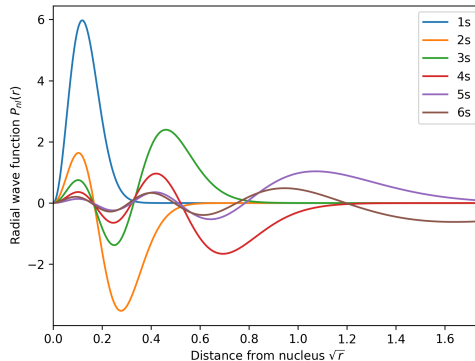
Types of correlation

Two main types of electron correlation have to be considered:

- **Static correlation** - Caused by degenerate HF energies
- **Dynamic correlation** - Arises from correlation of electron motions due to the repulsive Coulomb interaction

Static correlation - consider single-double-triple (SDT) substitutions or a multireference (MR) & Layzer complex

Dynamic correlation is harder... Introduce correlation orbitals

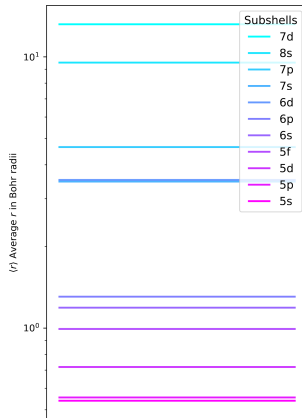


Core correlation

Core effects in Lr are strong, due to the ground state having a lone electron $[Rn] 5f^{14}7s^2 7p$

- The correlation orbitals should overlap with the valence orbitals
- Core orbitals close to the valence will affect energy separations

① *Stage One* (DHF) - Create wavefunction with DHF

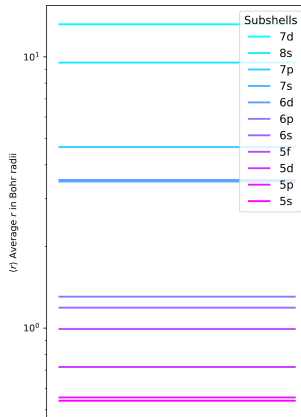


Core correlation

Core effects in Lr are strong, due to the ground state having a lone electron $[Rn] 5f^{14}7s^27p$

- The correlation orbitals should overlap with the valence orbitals
- Core orbitals close to the valence will affect energy separations

- 1 *Stage One* (DHF) - Create wavefunction with DHF
- 2 *Stage Two* (VV) - Add correlation orbitals by increasing the maximum principal quantum number by one. (One layer)

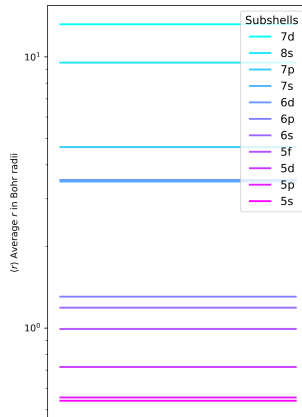


Core correlation

Core effects in Lr are strong, due to the ground state having a lone electron $[Rn] 5f^{14}7s^27p$

- The correlation orbitals should overlap with the valence orbitals
- Core orbitals close to the valence will affect energy separations

- 1 *Stage One* (DHF) - Create wavefunction with DHF
- 2 *Stage Two* (VV) - Add correlation orbitals by increasing the maximum principal quantum number by one. (One layer)
- 3 *Stage Three* (6sp/5sp) - Use the relativistic configuration interaction (RCI) to include effects from $\{6s, 6p\}$ subshells. Higher-order relativistic effects, such as the transverse photon interaction and leading QED corrections.

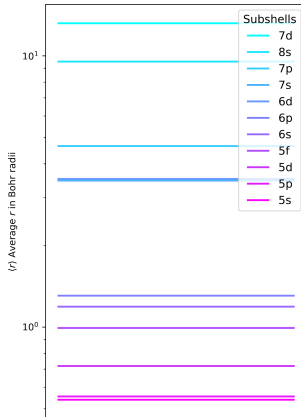


Core correlation

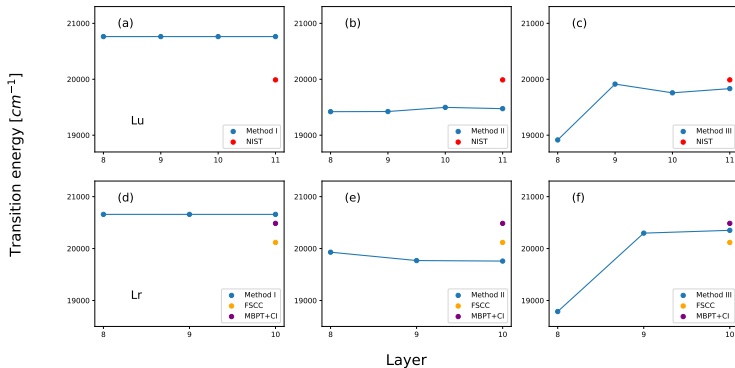
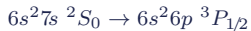
Core effects in Lr are strong, due to the ground state having a lone electron $[Rn] 5f^{14}7s^2 7p$

- The correlation orbitals should overlap with the valence orbitals
- Core orbitals close to the valence will affect energy separations

- 1 *Stage One* (DHF) - Create wavefunction with DHF
- 2 *Stage Two* (VV) - Add correlation orbitals by increasing the maximum principal quantum number by one. (One layer)
- 3 *Stage Three* (6sp/5sp) - Use the relativistic configuration interaction (RCI) to include effects from $\{6s, 6p\}$ subshells. Higher-order relativistic effects, such as the transverse photon interaction and leading QED corrections.
- 4 *Stage Four* (5df/4df) - Use RCI to include effects from $\{6s, 6p, 5d, 5f\}$ subshells. Include leading corrections as before



Graphical representation of transitions for Lu I and Lr I



Lu I and Lr I transition energies

Element	Levels J^P	Conf	Energy [cm^{-1}]					
			Method Two	Method Three	MR	Method Four	+ Static	NIST ⁴
Lu	$3/2^+$	$6s^2 5d$	0	0	0	0	0	0
Lu	$5/2^+$	$6s^2 5d$	590	1695	1619	1778	1702	1993
Lu	$1/2^-$	$6s^2 6p$	1334	3548	2358	3864	3864	4136
Lu	$3/2^-$	$6s^2 6p$	3765	6866	5686	7220	7230	7476
Lu	$1/2^+$	$6s^2 7s$	21353	23023	23523	23717	24217	24125
Lr	$1/2^-$	$7s^2 7p$	0	0	0	0	0	-
Lr	$3/2^+$	$7s^2 6d$	4907	3145	4853	-	-	-
Lr	$5/2^+$	$7s^2 6d$	7106	6047	7763	-	-	-
Lr	$3/2^-$	$7s^2 7p$	8133	8283	8540	8223	8480	-
Lr	$1/2^+$	$7s^2 8s$	20658	19930	22003	20351	20736	-
Lr	$3/2^+$	$7s^2 7d$	28543	27700	29922	28073	28607	-
Lr	$5/2^+$	$7s^2 7d$	28694	27949	30166	28277	28806	-

⁴A. Kramida and Y. Ralchenko, *NIST Atomic Spectra Database, NIST Reference Database 78, 1999.*

Transition rates

Element	Upper levels		Lower levels		Einstein A coefficient [s ⁻¹]				NIST ⁴
	J^P	Conf	J^P	Conf	Two	Three	Four	MR	
Lu	1/2 ⁺	6s ² 7s	1/2 ⁻	6s ² 6p	3.1 × 10 ⁷	4.1 × 10 ⁷	4.2 × 10 ⁷	3.2 × 10 ⁷	3.2 × 10 ⁷
Lu	1/2 ⁺	6s ² 7s	3/2 ⁻	6s ² 6p	5.2 × 10 ⁷	6.9 × 10 ⁷	7.2 × 10 ⁷	5.4 × 10 ⁷	4.9 × 10 ⁷
Element	Upper levels		Lower levels		Einstein A coefficient [s ⁻¹]				MBPT + CI ⁵
	J^P	Conf	J^P	Conf	Two	Three	Four	MR	
Lr	1/2 ⁺	7s ² 8s	1/2 ⁻	7s ² 7p	3.1 × 10 ⁷	3.4 × 10 ⁷	3.3 × 10 ⁷	3.3 × 10 ⁷	3.5 × 10 ⁷
Lr	3/2 ⁺	7s ² 7d	1/2 ⁻	7s ² 7p	5.0 × 10 ⁷	4.9 × 10 ⁷	4.6 × 10 ⁷	3.9 × 10 ⁷	6.1 × 10 ⁷
Lr	1/2 ⁺	7s ² 8s	3/2 ⁻	7s ² 7p	3.3 × 10 ⁷	3.3 × 10 ⁷	3.3 × 10 ⁷	2.8 × 10 ⁷	3.3 × 10 ⁷
Lr	3/2 ⁺	7s ² 7d	3/2 ⁻	7s ² 7p	9.5 × 10 ⁶	9.8 × 10 ⁶	9.7 × 10 ⁶	6.9 × 10 ⁶	1.2 × 10 ⁷
Lr	5/2 ⁺	7s ² 7d	3/2 ⁻	7s ² 7p	3.6 × 10 ⁷	5.2 × 10 ⁷	5.1 × 10 ⁷	3.9 × 10 ⁷	5.3 × 10 ⁷

Transition rates

Element	Upper levels		Lower levels		Einstein A coefficient [s ⁻¹]				NIST ⁴
	J^P	Conf	J^P	Conf	Two	Three	Four	MR	
Lu	1/2 ⁺	6s ² 7s	1/2 ⁻	6s ² 6p	3.1 × 10 ⁷	4.1 × 10 ⁷	4.2 × 10 ⁷	3.2 × 10 ⁷	3.2 × 10 ⁷
Lu	1/2 ⁺	6s ² 7s	3/2 ⁻	6s ² 6p	5.2 × 10 ⁷	6.9 × 10 ⁷	7.2 × 10 ⁷	5.4 × 10 ⁷	4.9 × 10 ⁷
Element	Upper levels		Lower levels		Einstein A coefficient [s ⁻¹]				MBPT + CI ⁵
	J^P	Conf	J^P	Conf	Two	Three	Four	MR	
Lr	1/2 ⁺	7s ² 8s	1/2 ⁻	7s ² 7p	3.1 × 10 ⁷	3.4 × 10 ⁷	3.3 × 10 ⁷	3.3 × 10 ⁷	3.5 × 10 ⁷
Lr	3/2 ⁺	7s ² 7d	1/2 ⁻	7s ² 7p	5.0 × 10 ⁷	4.9 × 10 ⁷	4.6 × 10 ⁷	3.9 × 10 ⁷	6.1 × 10 ⁷
Lr	1/2 ⁺	7s ² 8s	3/2 ⁻	7s ² 7p	3.3 × 10 ⁷	3.3 × 10 ⁷	3.3 × 10 ⁷	2.8 × 10 ⁷	3.3 × 10 ⁷
Lr	3/2 ⁺	7s ² 7d	3/2 ⁻	7s ² 7p	9.5 × 10 ⁶	9.8 × 10 ⁶	9.7 × 10 ⁶	6.9 × 10 ⁶	1.2 × 10 ⁷
Lr	5/2 ⁺	7s ² 7d	3/2 ⁻	7s ² 7p	3.6 × 10 ⁷	5.2 × 10 ⁷	5.1 × 10 ⁷	3.9 × 10 ⁷	5.3 × 10 ⁷

- $E_{\text{Total}} = E_{\text{Static}} + E_{\text{Dynamic}}$
- Assuming uncertainties are independent and random:
- $\delta E_{\text{Total}} = \sqrt{(\delta E)_{\text{Static}}^2 + (\delta E)_{\text{Dynamic}}^2}$

Comparison with previous theory

Levels		Energy [cm^{-1}]						
J^P	Conf	Our work	MBPT + CI ⁵	RCCSD ⁵	FSCC ⁶	CI + all order ⁷	MCDHF ⁸	MCDHF ⁹
$1/2^-$	$7s^27p$	0	0	0	0	0	0	0
$3/2^-$	$7s^27p$	8480	8606	8677	8413	8495	8138	7807
$1/2^+$	$7s^28s$	20736	20485	20533	20118	20253	20405	-
$3/2^+$	$7s^27d$	28607	28580	-	28118	-	-	-
$5/2^+$	$7s^27d$	28806	28725	-	28385	-	-	-

⁵E. V. Kahl et al., Physical Review A **104**, 052810 (2021).

⁶A. Borschevsky et al., The European Physical Journal D **45**, 115 (2007).

⁷V. A. Dzuba et al., Physical Review A **90**, 012504 (2014).

⁸S. Fritzsche et al., The European Physical Journal D **45**, 107 (2007).

⁹Y. Zou and C. Froese Fischer, Physical Review Letters **88**, 183001 (2002).

Summary

- Calculations were performed on Lu I and Lr I with good agreement with NIST and previous theory.
- Not properly including static correlation leads to a contracting effect of the separations
- $7s^2 8s \rightarrow 7s^2 7p = 20736 \pm 560 \text{cm}^{-1}$
- $7s^2 7d \rightarrow 7s^2 7p = 28607 \pm 672 \text{cm}^{-1}$.

Ab-initio multiconfigurational calculations of experimentally significant energy levels and transition rates in Lr I ($Z = 103$)

J. S. Andrews,^{1,2} J. Grumer,³ S. Fritzsche,^{1,2,4} A. Bondarev,^{2,4} P. Jönsson,⁵ and J. Bieroń⁶

¹Theoretisch-Physikalisches Institut, Friedrich-Schiller-Universität, 07743 Jena, Germany

²Helmholtz-Institut Jena, 07743 Jena, Germany

³Theoretical Astrophysics, Uppsala University, Uppsala, Sweden

⁴GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt, Germany

⁵Malmö University, Malmö, Sweden

Summary

- Calculations were performed on Lu I and Lr I with good agreement with NIST and previous theory.

- $7s^2 8s \rightarrow 7s^2 7p = 20736 \pm 560 \text{cm}^{-1}$
- $7s^2 7d \rightarrow 7s^2 7p = 28607 \pm 672 \text{cm}^{-1}$.

- Not properly including static correlation leads to a contracting effect of the separations

Lr I Outlook

- Further calculations could variationally incorporate static and dynamic correlation.
- To do this, new technologies would be needed such as non-orthogonal orbital sets or machine learning
- Hyperfine splittings of Lr I

Ab-initio multiconfigurational calculations of experimentally significant energy levels and transition rates in Lr I ($Z = 103$)

J. S. Andrews,^{1,2} J. Grumer,³ S. Fritzsche,^{1,2,4} A. Bondarev,^{2,4} P. Jönsson,⁵ and J. Bieroń⁶

¹Theoretisch-Physikalisches Institut, Friedrich-Schiller-Universität, 07743 Jena, Germany

²Helmholtz-Institut Jena, 07743 Jena, Germany

³Theoretical Astrophysics, Uppsala University, Uppsala, Sweden

⁴GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt, Germany

⁵Malmö University, Malmö, Sweden

Thank you for listening!

