Hyperfine-resolved laser spectroscopy of highly charged I\textsuperscript{7+} ions

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Outline

- Motivation
- Fine-structure and metastable states of Pd-like I$^{7+}$
- Hyperfine structure of Pd-like I$^{7+}$
- Brief of experimental observation and theoretical results
- Summary
Introduction

Hyperfine structure

Nuclear electron interactions induce particularly small splitting in atomic energy levels, defined as hyperfine structure.

For non-zero Nucleus spin \( F = |J-I| - |J+I| \)

\[ \text{nuclear spin} \quad \text{total angular momentum of e-} \]

HFS in Highly charged ion (HCl)

Highly charged ions (HCIs), enhanced hyperfine interactions owing to contracted electron clouds.
Motivation

- Hyperfine resolved spectroscopy of HCI’s play an important role in many studies

HFS in H-, He-, Li-, and Be-like ions have been widely performed

They have successfully contributed to
- Tests of relativistic and quantum electrodynamics (QED) atomic theories
- Investigations of nuclear properties

Toward the HCI clock (Good probe for fundamental physics)*

- Proposed atomic clocks are based on Hyperfine-structure resolved excitation (Viz. Ho^{14+}, Ir^{17+})
- Natural width of a clock transition involves hyperfine-mixing

Specific electron configuration with a 5s valence electron

**Ir$^{17+}$**

- $4f^{19}5s$
- $4f^{14}5s^2$
- $^3H_5$
- $^3F_4$
- $^3F_3$
- $^3F_2$
- $^1S_0$
- E1-hfs clock, $\lambda \sim 2000$ nm
- M2 clock, $\lambda \sim 300$ nm

**Ho$^{14+}$**

- $4f^{6}5s$
- $4f^{5}5s^2$
- $^4P_{3/2}(11)$
- $^6F_{3/2}(10)$
- $^6F_{5/2}(9)$
- $^6F_{7/2}(6)$
- $^6H_{15/2}(5)$
- $^6H_{13/2}(4)$
- $^6H_{11/2}(3)$
- $^6H_{9/2}(2)$
- $^6H_{7/2}(1)$

**TABLE III.** Magnetic-dipole and electric-quadrupole hfs constants $A$ and $B$ for the ground and clock states of $^{165}$Ho$^{14+}$.

<table>
<thead>
<tr>
<th>State</th>
<th>A (GHz)</th>
<th>B (GHz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4f^{6}5s$</td>
<td>96.5</td>
<td>0</td>
</tr>
<tr>
<td>$4f^{5}5s^2$</td>
<td>3.53</td>
<td>−6.04</td>
</tr>
</tbody>
</table>


Large HFS constant ??


ASOS14, Paris, July 10-13, 2023
System of Interest  Pd-like $^{127}$I$^{7+}$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

$4d^95s^1$

Collisional radiative Model calculations

<table>
<thead>
<tr>
<th>Label</th>
<th>Level</th>
<th>$\tau$ (s)</th>
<th>$\rho$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$\left(4d_{5/2}^{-1}5s_{1/2}\right)_{J=3}$</td>
<td>$3.6 \times 10^3$</td>
<td>17.64</td>
</tr>
<tr>
<td>B</td>
<td>$\left(4d_{3/2}^{-1}5s_{1/2}\right)_{J=1}$</td>
<td>$3.3 \times 10^{-2}$</td>
<td>1.92</td>
</tr>
<tr>
<td>C</td>
<td>$\left(4d_{5/2}^{-1}4f_{3/2}\right)_{J=6}$</td>
<td>$3.2 \times 10^{-4}$</td>
<td>0.13</td>
</tr>
<tr>
<td>D</td>
<td>$\left(4d_{5/2}^{-1}4f_{5/2}\right)_{J=5}$</td>
<td>$1.9 \times 10^{-4}$</td>
<td>0.13</td>
</tr>
<tr>
<td>E</td>
<td>$\left(4d_{5/2}^{-1}4f_{5/2}\right)_{J=5}$</td>
<td>$2.3 \times 10^{-4}$</td>
<td>80.11</td>
</tr>
</tbody>
</table>

Kimura et al., PRA102, 032807 (2020)
Hyperfine splitting (Pd-like- $^{127}$I$^{7+}$)

$4d^{10}5s^1$

Nuclear Spin, $I = 5/2$

Angular momentum $J = 3, 2$

$F = |I + J|$ to $|I - J|$

$\bullet$ What is the order of the hyperfine splitting?

$\bullet$ Does hyperfine mixing will change the lifetime of the metastable states?
Multi configuration Dirac-Fock (MCDF) calculations using GRASP2018*

- Core–core and core–valence correlations with the inner orbitals were also included.
- This active space treatment led to 3,300,000 jj-coupled configurations.

\[
\begin{align*}
\text{DF} &= \{3s^2 3p^6 4s^2 4p^6 4d^{10}, 3s^2 3p^6 4s^2 4p^6 4d^9 5s^1 \}, \\
\text{AS1} &= \text{DF} + \{5p, 5d, 5f, 5g\}, \\
\text{AS2} &= \text{AS1} + \{6s, 6p, 6d, 6f, 6g, 6h\}, \\
\text{AS3} &= \text{AS2} + \{7s, 7p, 7d, 7f, 7g, 7h\}, \\
\text{AS4} &= \text{AS3} + \{8s, 8p, 8d, 8f, 8g, 8h\}.
\end{align*}
\]

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<table>
<thead>
<tr>
<th></th>
<th>Theory</th>
<th>MCDF</th>
<th>Breit</th>
<th>QED</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A_{\text{hfs}}) [GHz]</td>
<td>10.39 (± 0.05)</td>
<td>10.41</td>
<td>(-1.7 \times 10^{-2})</td>
<td>+ 3.3 \times 10^{-3}</td>
</tr>
<tr>
<td>(B_{\text{hfs}}) [GHz]</td>
<td>2.32 (± 0.02)</td>
<td>2.37</td>
<td>(-4.3 \times 10^{-2})</td>
<td>+ 4.0 \times 10^{-4}</td>
</tr>
<tr>
<td>(A'_{\text{hfs}}) [GHz]</td>
<td>15.33 (± 0.03)</td>
<td>15.45</td>
<td>(-1.2 \times 10^{-1})</td>
<td>+ 7.5 \times 10^{-3}</td>
</tr>
<tr>
<td>(B'_{\text{hfs}}) [GHz]</td>
<td>2.02 (± 0.01)</td>
<td>2.05</td>
<td>(-2.8 \times 10^{-2})</td>
<td>+ 3.0 \times 10^{-4}</td>
</tr>
<tr>
<td>(k_0) [cm(^{-1})]</td>
<td>17616 (± 22)</td>
<td>18016</td>
<td>(-418)</td>
<td>+ 18</td>
</tr>
</tbody>
</table>

\[
\Delta E = \frac{AK}{2} + B \frac{3K(K+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)},
\]

with \(K = F(F+1) - I(I+1) - J(J+1)\).

*C. Froese Fischer, et. al., Computer Physics Communications 2019, 237, 184*
**Hyperfine induced transition Rates Mixing splitting (Pd-like- \( ^{127}\text{I}^{7+} \))**

Specifications

Vacuum : $\sim 10^{-9}$ Pa
Beam energy : 50-2000 eV
Beam current : 1~20 mA
Magnetic field : 0.03 - 0.2 T
**I^7+ spectroscopy concept: Plasma-assisted laser spectroscopy**

**Specifications**

- **Vacuum**: ~10^{-9} Pa
- **Beam energy**: 50-1000 eV
- **Beam current**: 1-20 mA
- **Magnetic field**: 0.03 - 0.2 T

**Uranium Emission Current** (UEC compact EBIT (CoBIT))

- Collector
- DT
- E-Gun

**Collisional and radiative processes**

- Laser excitation (M1) 567 nm, 10 ns pulse
- LIF (E2)

- M1 selection rule
  \[ \Delta F = \pm 1, 0 \]
  \[ \perp : \Delta M_F = 0 \]

- Calculated by Grasp

**Beam energy and current**

- Beam energy: 50-1000 eV
- Beam current: 1-20 mA

**Vacuum**

- ~10^{-9} Pa

**Magnetic field**

- 0.03 - 0.2 T
Can we resolve the hyperfine-structure??
**Experiment**

YAG laser

Dye laser

Polarizer

λ/2

Vacuum chamber

CoBIT

Power meter

Polarizer: λ/2

Accuracy: 0.02 cm⁻¹

Line width: ~0.03 cm⁻¹

**Diagram Details:**

- YAG laser
- Dye laser
- Polarizer
- Vacuum chamber
- CoBIT
- Power meter
- Grating
- Wavemeter
- Position analyzer
- Amp.
- PSD
- DG
- MCS
- FG
- Trigger
- LIF timing

**Notations and Formulas:**

- $\Delta m_F = 0$
- $\lambda = 532$ nm
- $\perp 10$ ns pulse
- $M_1$ and $E_2$
- $25$ nm
- $\tau \sim 5 \mu$s
- $4d^{10}$ (Ground)
- $4d_3^{1/2} 5s_{1/2}, J = 2$
- $4d_5^{5/2} 5s_{1/2}, J = 3$
- $567$ nm

**Data Points:**

- Continuous LIF time range: 13
- ASOS14, Paris, July 10-13, 2023

**Graphs:**

- Time-resolving graph
- LIF time range (10 µs)
- Wavelength in vacuum measured with the EUV spectrometer [nm]

**Additional Information:**

- Dye laser
- SHG (532 nm)
- 100 Hz Repetition
- 7 mJ

**Figures and Equations:**

- $J = 2 - 4d^{10}$
- $J = 3 - 4d^{10}$
- $J = 1 - 4d^{10}$
- $4d_3^{1/2} 5s_{1/2}, J = 2 - 4d^{10}$
- $4d_5^{5/2} 4f_{7/2}, J = 1 - 4d^{10}$
Wavelength spectrum for the LIF signal

**Good Agreement!**

<table>
<thead>
<tr>
<th></th>
<th>Th. (GRASP)</th>
<th>Exp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_{J=3 \rightarrow J=2}$</td>
<td>17616 cm$^{-1}$</td>
<td>17633.67 (±0.05) cm$^{-1}$</td>
</tr>
<tr>
<td>$A_{hfs}$ (J=3)</td>
<td>10.39 GHz</td>
<td>10.3 (±0.6) GHz</td>
</tr>
<tr>
<td>$B_{hfs}$ (J=3)</td>
<td>2.32 GHz</td>
<td>2.9 (±2.1) GHz</td>
</tr>
<tr>
<td>$A_{hfs}$ (J=2)</td>
<td>15.33 GHz</td>
<td>15.8 (±0.6) GHz</td>
</tr>
<tr>
<td>$B_{hfs}$ (J=2)</td>
<td>2.02 GHz</td>
<td>1.5 (±1.6) GHz</td>
</tr>
</tbody>
</table>

Where is the resonance wavelength ??

Grasp calculation

<table>
<thead>
<tr>
<th>Term</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dirac-Fock (DF)</td>
<td>18241.15</td>
</tr>
<tr>
<td>Valence-valence correlation (VV)</td>
<td>-481.30</td>
</tr>
<tr>
<td>Core-valence correlation (CV)</td>
<td>326.82</td>
</tr>
<tr>
<td>Core-core correlation (CC)</td>
<td>-70.62</td>
</tr>
<tr>
<td>Breit interaction (Breit)</td>
<td>-418.24</td>
</tr>
<tr>
<td>self-energy (SE)</td>
<td>17.34</td>
</tr>
<tr>
<td>vacuum polarization (VP)</td>
<td>0.80</td>
</tr>
<tr>
<td>Total transition energy</td>
<td>17616.03±22 cm(^{-1})</td>
</tr>
</tbody>
</table>
We also measured the microsecond-order lifetime of \((4d^95s^1)_{J=2}\) in Pd-like I\(^{7+}\) using pulsed laser excitation from a metastable state.

While the experimental lifetime of this state has the potential to be a benchmark for developing reliable atomic structure calculations of relativistic many-electron systems with \(d\) electrons, it is generally difficult to measure such short lifetimes.

### TABLE 1. Summary of the experimental and theoretical lifetime \(\tau\) with calculated individual transition probabilities. The theoretical values were calculated by employing the active space set AS4 and include the RCI correction.

<table>
<thead>
<tr>
<th>Decay channel</th>
<th>Experiment</th>
<th>Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A_{1,2}) (s(^{-1}))</td>
<td>((4d^{10})_{J=0})</td>
<td>2.32 \times 10^3</td>
</tr>
<tr>
<td>(A_{M1,J=3}) (s(^{-1}))</td>
<td>((4d^{9}5s)_{J=3})</td>
<td>4.95 \times 10^1</td>
</tr>
<tr>
<td>(A_{M1,J=2}) (s(^{-1}))</td>
<td>((4d^{9}5s)_{J=2})</td>
<td>3.06 \times 10^0</td>
</tr>
<tr>
<td>(A_{M1,J=1}) (s(^{-1}))</td>
<td>((4d^{9}5s)_{J=1})</td>
<td>3.52 \times 10^{-1}</td>
</tr>
<tr>
<td>(A_{\text{total}}) (s(^{-1}))</td>
<td>(2.32(\pm 0.07_{\text{stat}} \pm 0.01_{\text{sys}}) \times 10^6)</td>
<td>2.32 \times 10^6</td>
</tr>
<tr>
<td>(\tau) ((\mu)s)</td>
<td>(4.31(\pm 0.14_{\text{stat}} \pm 0.02_{\text{sys}}))</td>
<td>4.31</td>
</tr>
</tbody>
</table>

FIG. 3. Bottom: Experimentally observed LIF decay profile. The red line represents the fitting result. Top: Residuals of the experimental plots from the fitting line. Error bars reflect Poisson counting statistics.
Theoretical transition probability using GRASP2018*

DF = \{3s^2 3p^6 4s^2 4p^6 4d^{10}, 3s^2 3p^6 4s^2 4p^6 4d^9 5s^1 \},
AS1 = DF + \{5p, 5d, 5f, 5g\},
AS2 = AS1 + \{6s, 6p, 6d, 6f, 6g, 6h\},
AS3 = AS2 + \{7s, 7p, 7d, 7f, 7g, 7h\},
AS4 = AS3 + \{8s, 8p, 8d, 8f, 8g, 8h\}.

SD excitation from 3s, 3p, 3d, 4s, 4p, 4d

Active space set dependence of the E2 transition line strength (in atomic units) in the MCDF calculation without the RCI correction. The thin black lines represent the experimental uncertainty (1σ).

* C. Froese Fischer, et. al., Computer Physics Communications 2019, 237, 184
Summary

- We have demonstrated laser spectroscopy of forbidden transitions between metastable states of HCl isotopes stored in an EBIT by employing Pd-like $^{127}$I$^7^+$.  

- The laser excitation spectrum of the HCl isotopes in a quasi-Zeeman-free low magnetic field revealed distinct hyperfine structures.

- Even though the transition observed in this study is not a proposed HCI clock candidate, the building of a benchmark to understand hyperfine structures in many-electron HCl isotopes.
