Selective laser ionization of odd-mass number isotopes 
*(odd-mass selection)*

for the partitioning of palladium and zirconium

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Reduction and resource recycling of high-level radioactive wastes through nuclear transmutation

High-level radioactive wastes are transformed into low and mid-level ones and a portion of the rare metals is reused.

http://www.jst.go.jp/impact/en/program/08.html
Our mission in ImPACT program

Develop efficient extraction technique of LLFP isotopes ($^{93}\text{Zr}^*$, $^{107}\text{Pd}^*$) aiming at both the nuclear transmutation and recycling.
Isotope separation vs. odd-mass selection

In the case of palladium (Pd)

Mixture of palladium isotopes in HLW

Isotope separation
Wavelength of laser

107

w/ ultra-high resolution laser
No precise spectroscopic data

Odd-mass selection
Non-zero nuclear spin

105
107

Easy

102 104 105 106 107 108 110
220 375 341 212 146 48 g / ton*

*JAERI-M 91-147
Difference in the electronic state structure of palladium between even-mass \((I=0)\) and odd-mass \((I\neq 0)\) isotopes

**Even-mass number isotopes \((I = 0)\)**

- **4d\(^9\)5d**
  - \(J = 0\)
  - \(m_j = -1, 0, +1\)

- **4d\(^9\)5p**
  - \(J = 1\)
  - \(m_j = -1, 0, +1\)

**Odd-mass number isotopes\(^{105, 107}\)Pd \((I = 5/2)\)**

- **4d\(^9\)5d**
  - \(J = 0\)
  - \(m_f = -7/2, -5/2, -3/2, -1/2, +1/2, +3/2, +5/2, +7/2\)
  - \(F = J + I\)

\(F = 5/2\)

- **4d\(^9\)5p**
  - \(J = 1\)
  - \(m_f = -3/2, -1/2, +1/2, +3/2, +5/2, +7/2\)
  - \(F = 3/2, 5/2, 7/2\)

\(F = 5/2\)
Selection rules for electronic transition absorption of photons

\[ \Delta J = 0, \pm 1 \]

Total angular momentum \( F = J + I \)

☆ Linear polarization

Rule
\[ \Delta m_J = 0 \]
\[ \Delta m_F = 0 \]

Nuclear spin
\[ I = 0 \]

Easy to maintain

Nuclear spin
\[ I \neq 0 \]

Not easy to maintain

☆ Circular polarization

Rule
\[ \Delta m_J = \pm 1 \]
\[ \Delta m_F = \pm 1 \]

Nuclear spin
\[ I = 0 \]

+1 for LHC and -1 for RHC.

Nuclear spin
\[ I \neq 0 \]

We need to choose proper combination of electronic states of particular \( J \) to realize selective excitation and ionization.
Original scheme proposed by Hao-Lin Chen (1980)

2-LHC lasers + ionization laser: 3 lasers

Only odd-mass isotopes absorb the 2nd laser photon

\[ m_J = -1 \quad 0 \quad +1 \quad m_F = -\frac{7}{2} \quad -\frac{5}{2} \quad -\frac{3}{2} \quad -\frac{1}{2} \quad +\frac{1}{2} \quad +\frac{3}{2} \quad +\frac{5}{2} \quad +\frac{7}{2} \]

Even-mass isotopes \((I = 0)\)

Odd-mass isotopes \(^{105},^{107}\)Pd \((I = 5/2)\)
Drawbacks of the original scheme

*For selective excitation*

- Using two circularly polarized lasers
  - Not easy to maintain polarization
  - Not suitable for multi-pass optics

*For ionization*

- Non-resonant ionization  Low efficiency

*As for the Cost*

- Totally 3 lasers for selective ionization
  - High initial and maintenance costs
We have developed 2-laser scheme.

For selective excitation
  • Using two // linearly polarized lasers
    Easy to maintain polarization
    Suitable for multi-pass optics

For ionization
  • Resonant ionization = High efficiency

As for the Cost
  • Reduced number of lasers to 2
    Less initial and maintenance costs
2-laser scheme

Original scheme (2-step excitation + ionization)

2-laser scheme (2-step excitation/ionization)

Ionization laser

autoionizing states

Even + Odd

Even - odd separation

Even + Odd

Even + Odd

Odd

Even + Odd

Even + Odd

Ionization continuum

Need to confirm if negligible or not.

Non-resonant ionization

Even - odd separation

Even + Odd

Even + Odd

Even + Odd

Resonant ionization

Even + Odd

Ionization limit

Spectroscopic investigation of autoionizing states
Selective (//) and non-selective (⊥) excitations

\[ 4d^9(2D_{3/2})nd \ (J = 1) \]

Selectivity

Non-selectivity
Selectivity check

\[ \text{Selectivity check} \]

105\text{\textsuperscript{Pd}}^+ > 99.7 \%

Non-selective

Selective

The result suggests transition to ionization continuum is negligible.
Zirconium: Tuning $\omega_3$ in search of $J=0$ state

$J = 2 \Rightarrow 1 \Rightarrow 1 \Rightarrow 0 \Rightarrow$ ionization

Selective excitation with 3-// linearly polarized lasers

1064 nm

$\omega_3$ scan

Reported by Niki

$^{91}\text{Zr}^+ > 98.6\%$

$\beta > 575$

30 times larger intensity than the previous report.

Dr. Niki’s line
Effort to increase ion yield (Pd)

Simple multi-pass optics

Coaxial 2 laser beams

Vapor of Pd \(1.1 \times 10^{-5} \text{Vs} @ 12.0 \text{Ås}^{-1} = 1.4 \times 10^{12} \text{ions/pulse}\)

- 0.21 g/day @ 10 kHz
- 77 g/year @ 10 kHz

10-pass

Single-pass
Road to practical realization

• High power lasers
• Large volume multi-pass optics

High-level nuclear waste (HLW) 20 ton/year
Pd 27 kg/year

Our value 77 g/year

The difference will be overcome in the near future.
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