



TASCA 16

15th Workshop on
Recoil Separator for Superheavy Element Chemistry

Measurements of the first ionization potentials of the heaviest actinides

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Contents

1) Measurements of the first ionization potential (IP_1) of the heavy actinides at JAEA

2) Recent activities at JAEA

- Adsorption behavior of lawrencium (Lr, $Z=103$) on Ta surface**

IP₁ measurements of heavy elements



LETTER

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Measurement of the first ionization potential of lawrencium, element 103

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The chemical properties of an element are primarily governed by the configuration of electrons in the valence shell. Relativistic effects influence the electronic structure of the periodic table, and the seventh row—including the configurations^{1,2}. Atomic s orbitals exhibit relativistic effects, whereas p_{3/2}, d ground-state configurations of lighter elements in the same row (IP₁) is a measure of the energy required to remove an electron from the atom. The spallation reaction of uranium¹⁴. IP₁ values of heavy elements with Z ≥ 100, however, could not be determined experimentally, because

T. K. Sato *et al.*

Nature 520 (Apr.9) (2015) 209–211.

Electronic configuration of heavy elements influenced by strong rel. eff.



Experimental determination of the first ionization potential (IP₁)

Surface ionization

**Low production rate and/or
short half-lives**

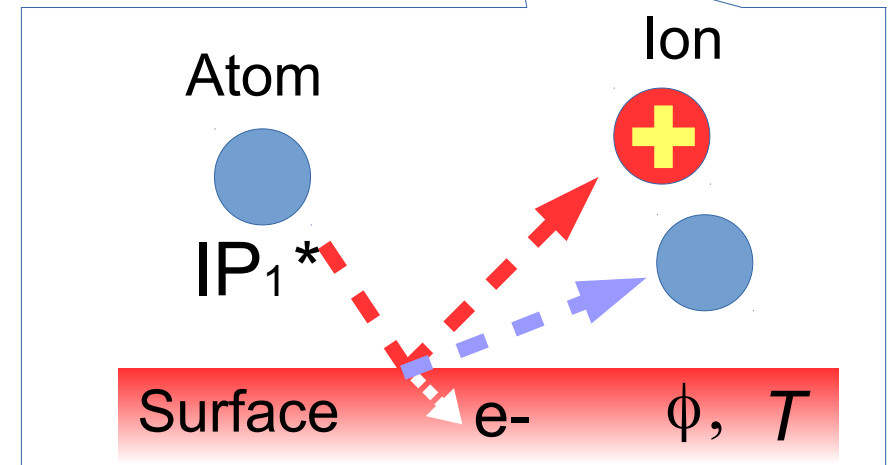
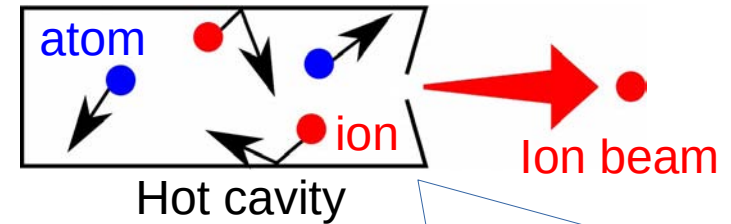


**A NEW IP_1 measurement
method based on surface
ionization process**

Surface ionization

**Low production rate and/or
short half-lives**

**A NEW IP_1 measurement
method based on surface
ionization process**



Surface ionization

Saha-Langmuir eq.

$$I_{\text{eff}} = \frac{N \exp((\phi - IP_1^*)/kT)}{1 + N \exp((\phi - IP_1^*)/kT)}$$

Effective $IP_1(IP_1^*)$: $IP_1^* = IP_1 - kT \ln(Q_i/Q_o)$

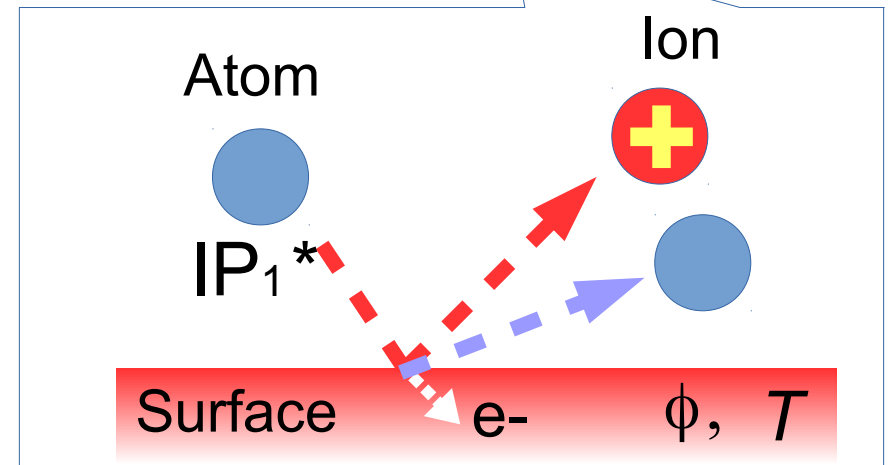
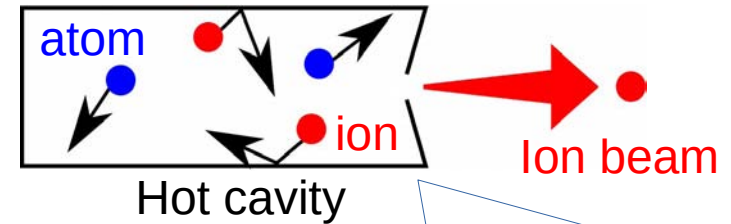
I_{eff} : ionization efficiency

ϕ : work function [eV]

T : surface temperature [K]

Q_i and Q_o : partition functions of ion and atom

IP_1 : The first ionization potential



Surface ionization

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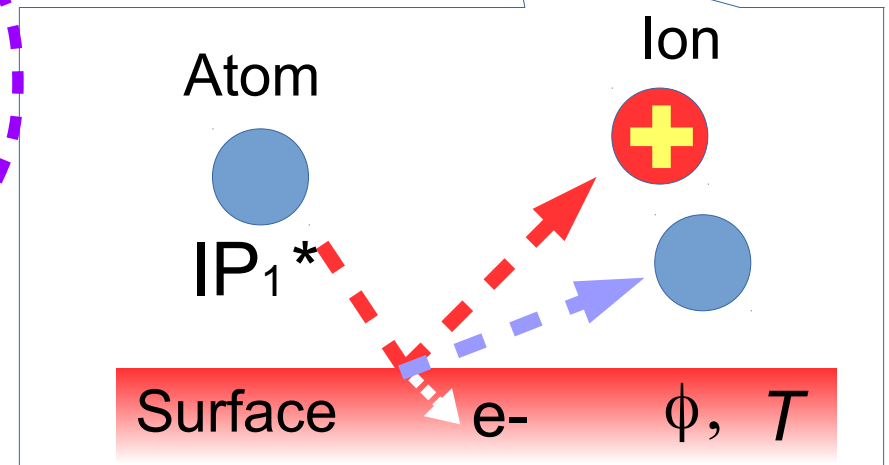
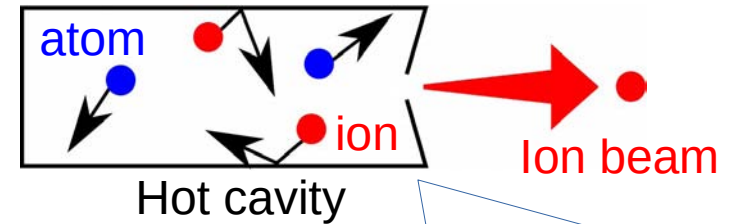
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Ionization eff. (I_{eff}) \rightarrow **Effective IP_1 (IP_1^*)**

Surface ionization

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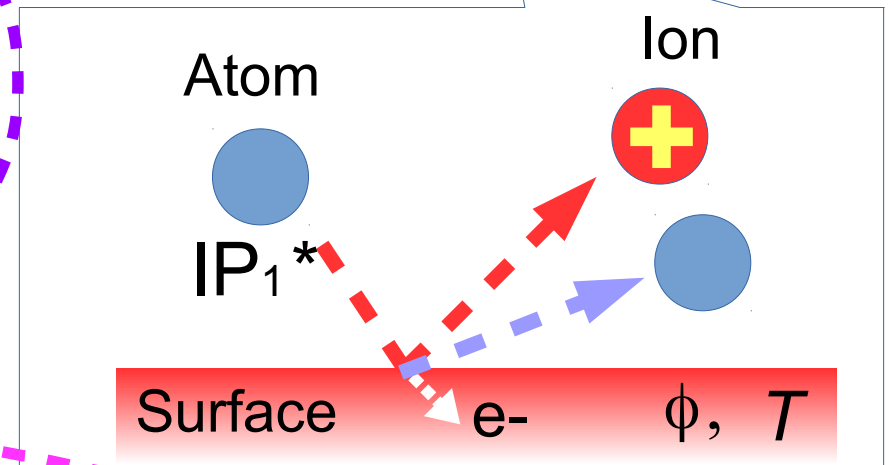
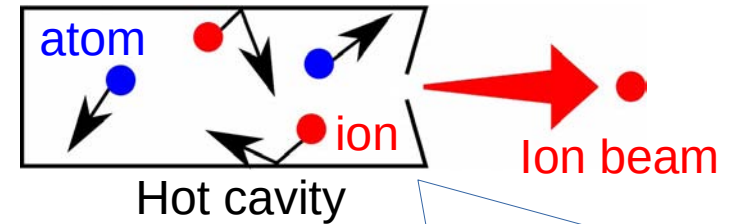
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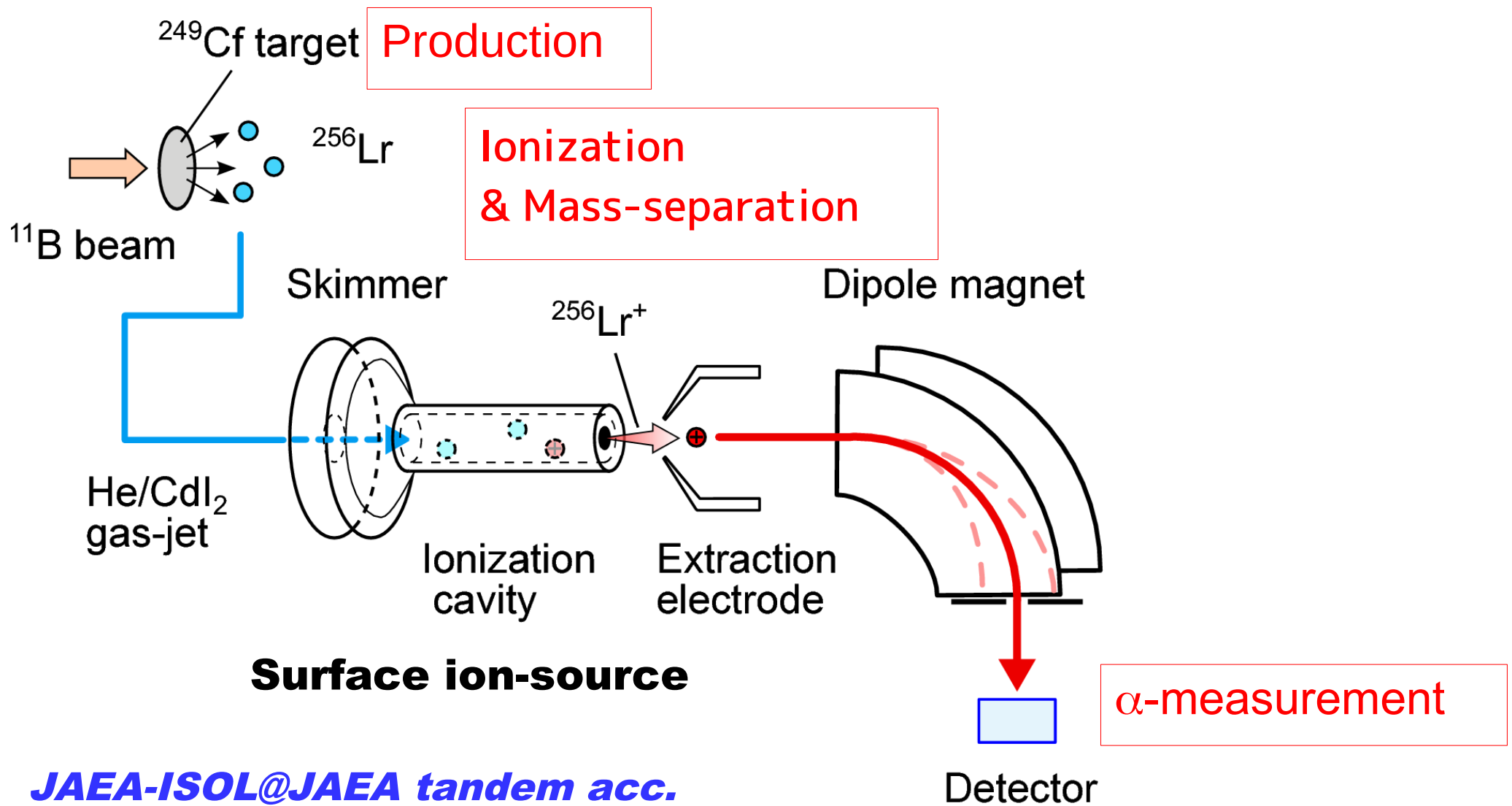
Q_i and Q_o : partition functions of ion and atom

IP_1 : The first ionization potential



Ionization eff. (I_{eff}) \rightarrow **Effective IP_1 (IP_1^*)** \rightarrow **IP_1** 🚩

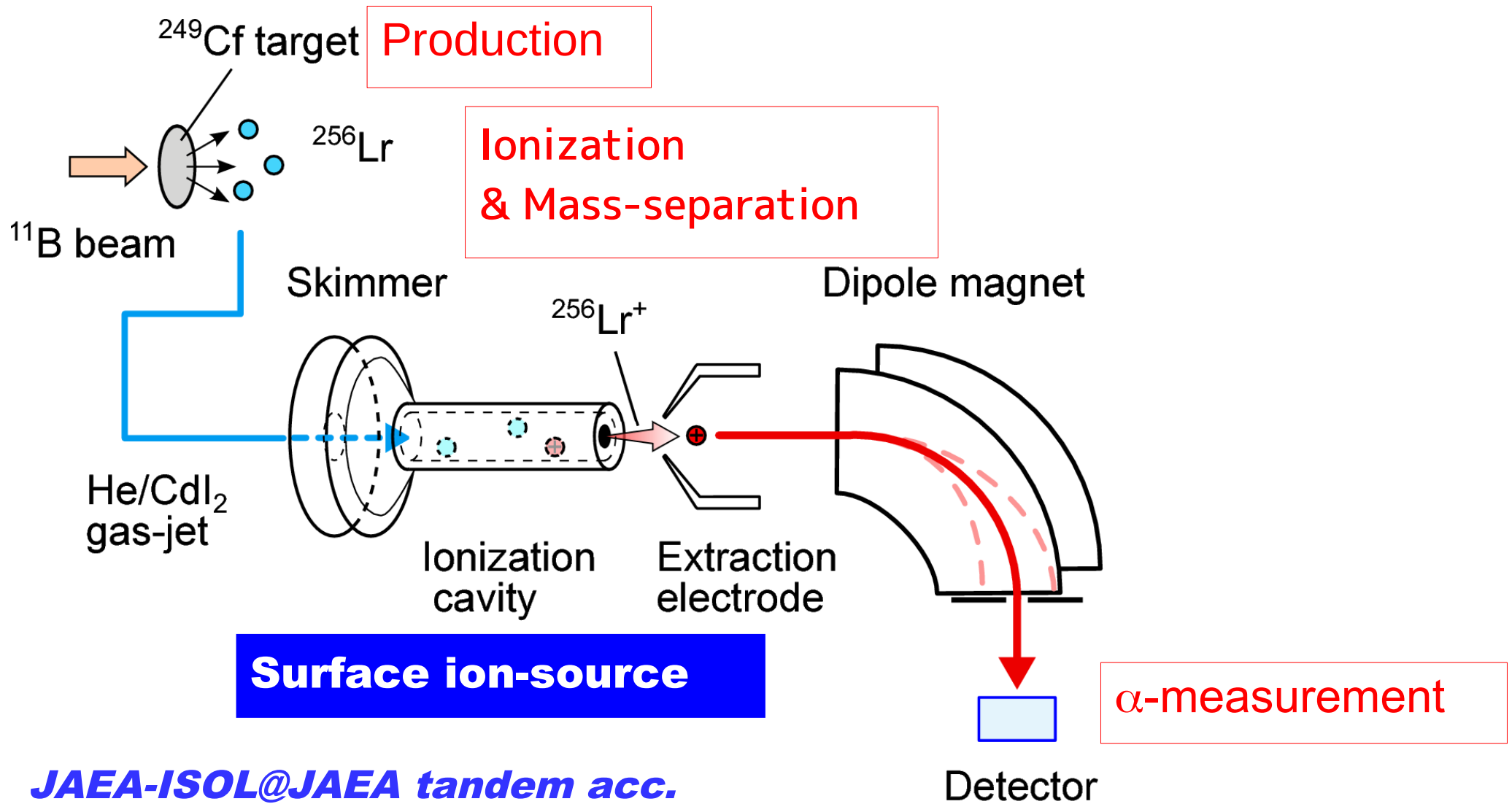
Experimental setup



JAEA-ISOL@JAEA tandem acc.

[1] T.K.Sato et al JRNC, 303, 1253-1257 (2015).

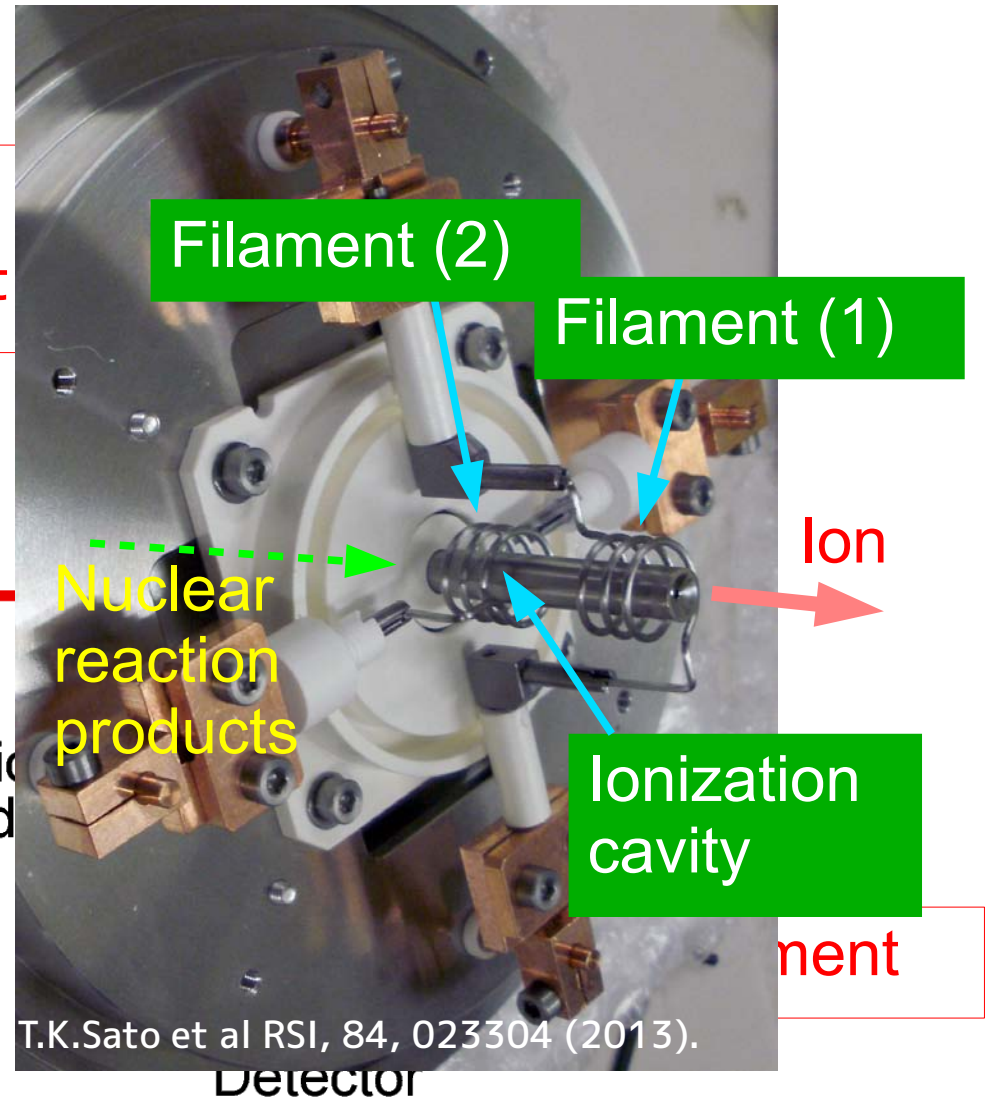
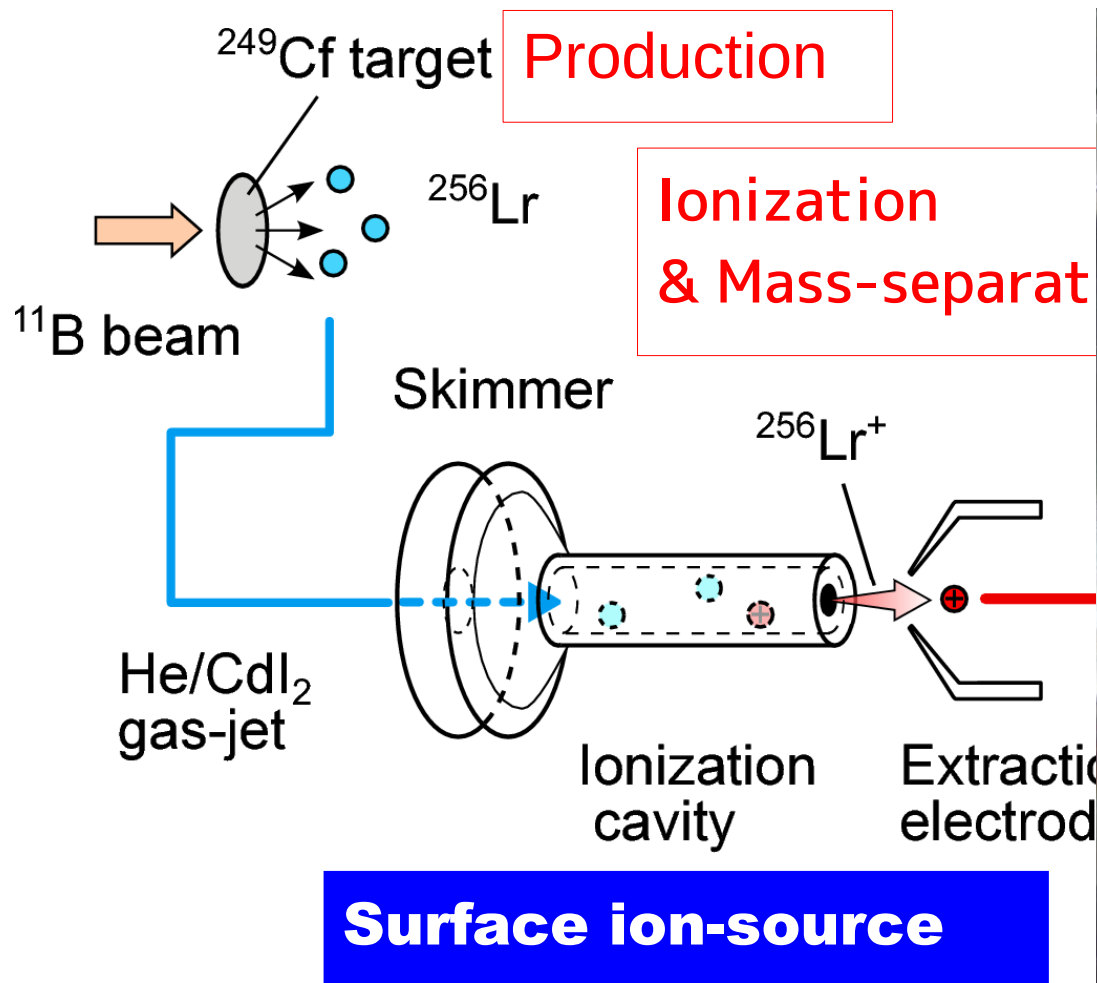
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Experimental setup



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IP₁ measurement of Lr

Isotopes : ⁸⁰Rb, ^{142,143}Eu, ¹⁴³Sm,
¹⁴⁸Tb, ^{153,154}Ho, ¹⁵⁷Er,
¹⁶²Tm, ¹⁶⁵Yb, & ¹⁶⁸Lu

Beam : ¹¹B(67.9 MeV)

Targets : ¹³⁶Ce, ¹⁴¹Pr, ¹⁴²Nd, ¹⁴⁷Sm,
Eu, ¹⁵⁶Gd, ¹⁵⁹Tb,
¹⁶²Dy, and Ge

Ionization temp.: 2700K



S-L eq.

$$I_{\text{eff}} = \frac{N \exp((\phi - \text{IP}_1^*)/kT)}{1 + N \exp((\phi - \text{IP}_1^*)/kT)}$$

in the present system

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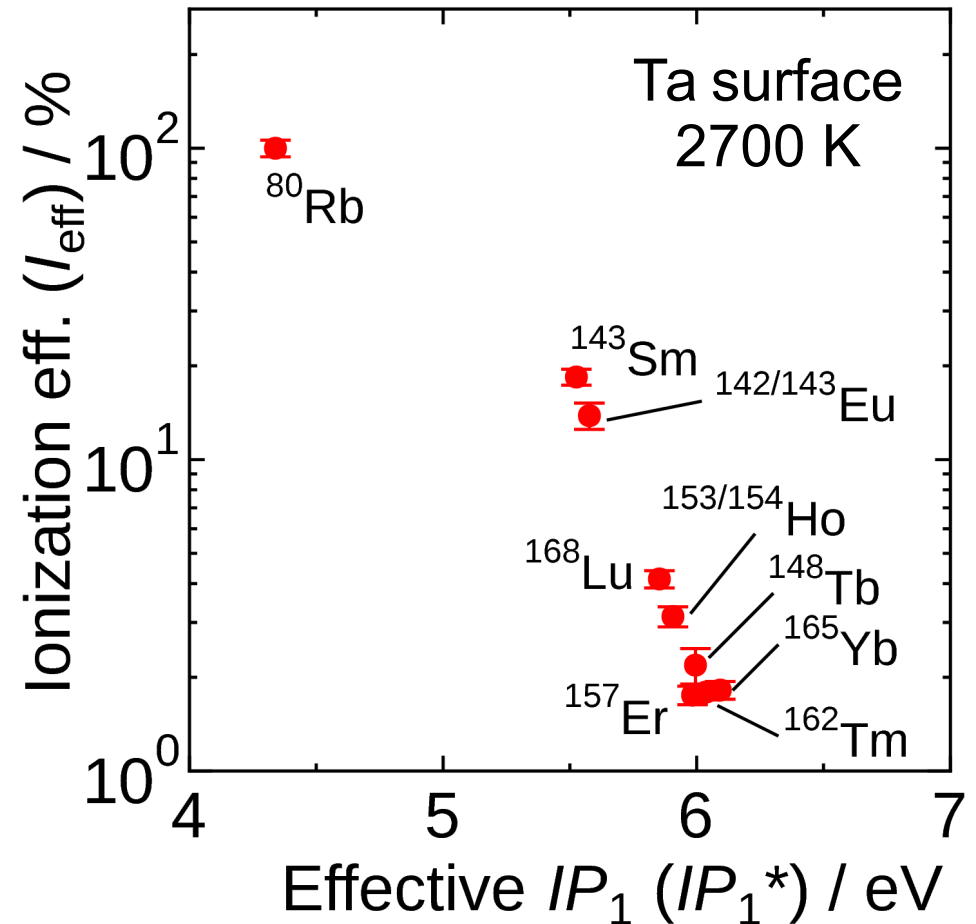
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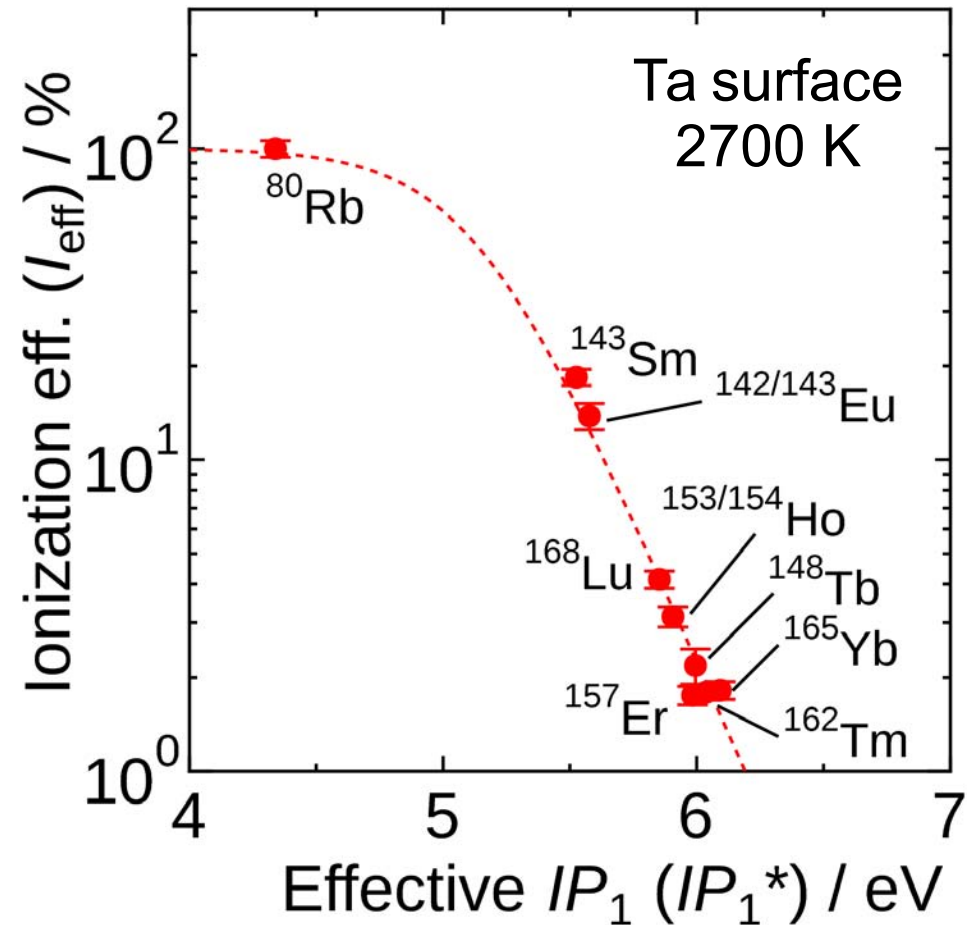
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S-L eq.

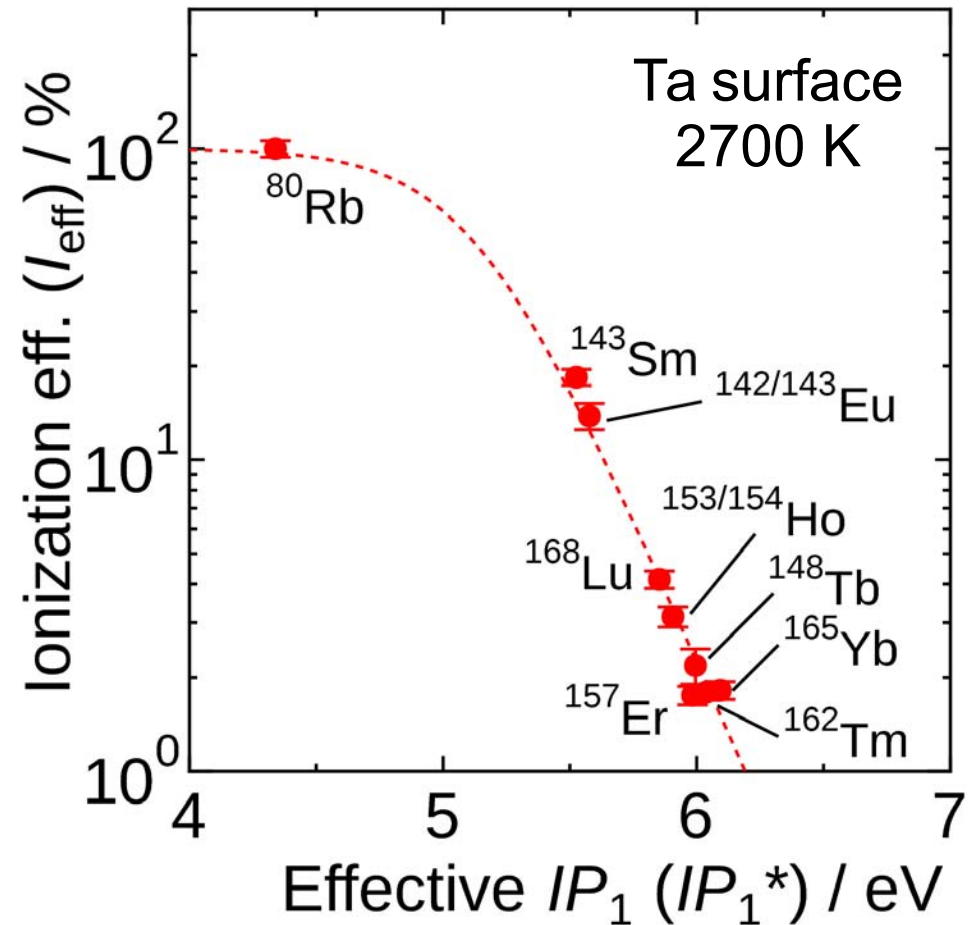
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in the present system



IP₁ measurement of Lr

Isotope: ²⁵⁶Lr ($T_{1/2} = 27$ s)
Beam : ¹¹B (67.9 MeV)
Target : ²⁴⁹Cf (260 $\mu\text{g}/\text{cm}^2$)
Ionization temp.: 2700K

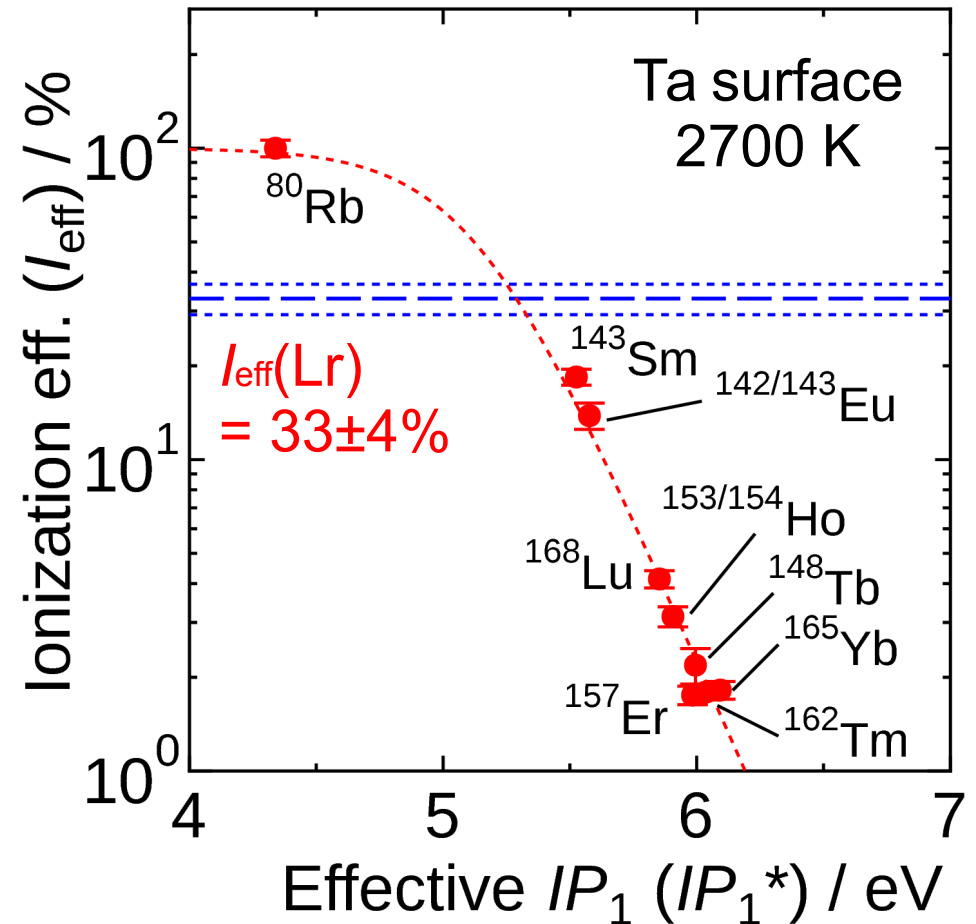


IP₁ measurement of Lr

Isotope: ²⁵⁶Lr ($T_{1/2} = 27$ s)
Beam : ¹¹B (67.9 MeV)
Target : ²⁴⁹Cf (260 $\mu\text{g}/\text{cm}^2$)
Ionization temp.: 2700K



$I_{\text{eff}}(\text{Lr}) = 33 \pm 4\%$
in the present system

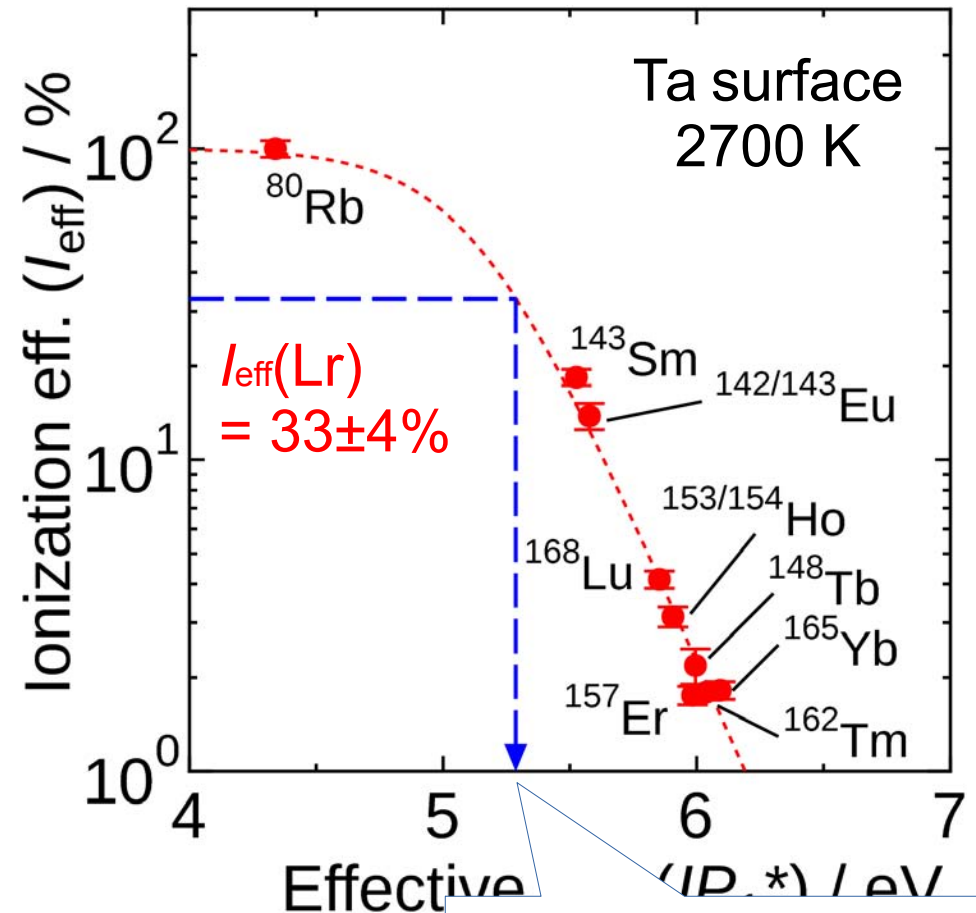


IP₁ measurement of Lr

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Target : ²⁴⁹Cf (260 μg/cm²)
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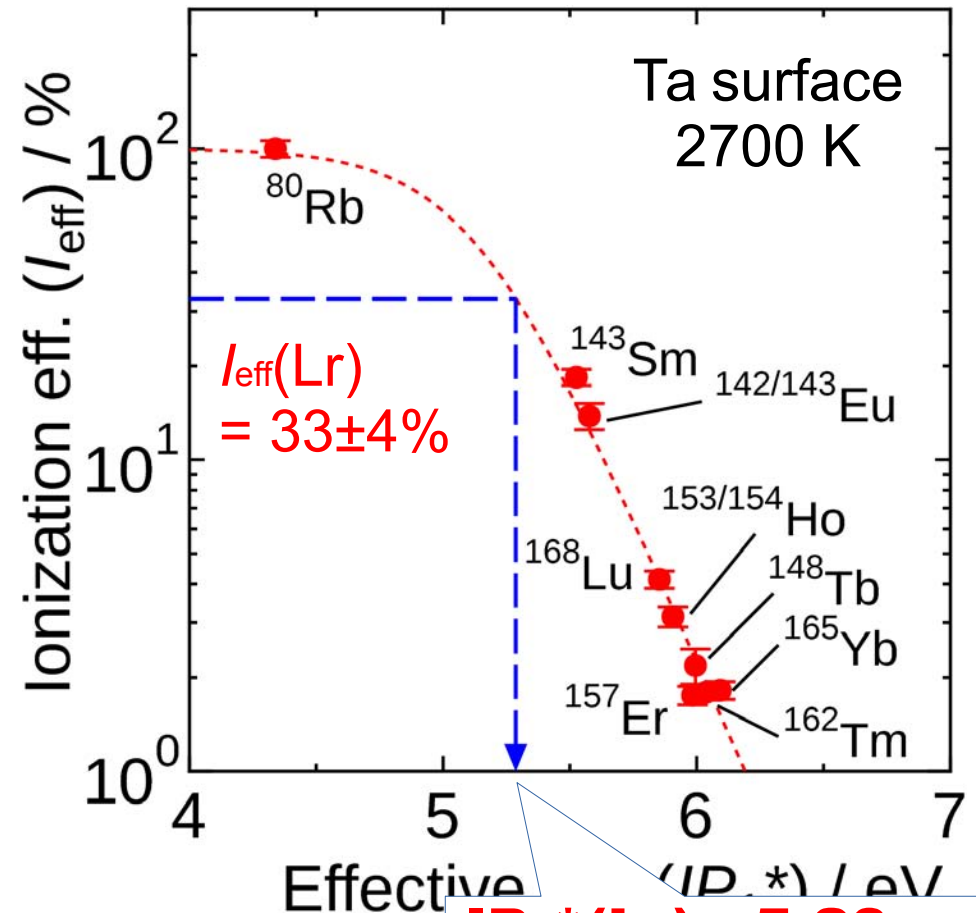


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$I_{\text{eff}}(\text{Lr}) = 33 \pm 4\%$
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$IP_1^*(\text{Lr}) = 5.29$



$IP_1(\text{Lr}) = 4.96$

IP₁ measurement of Lr

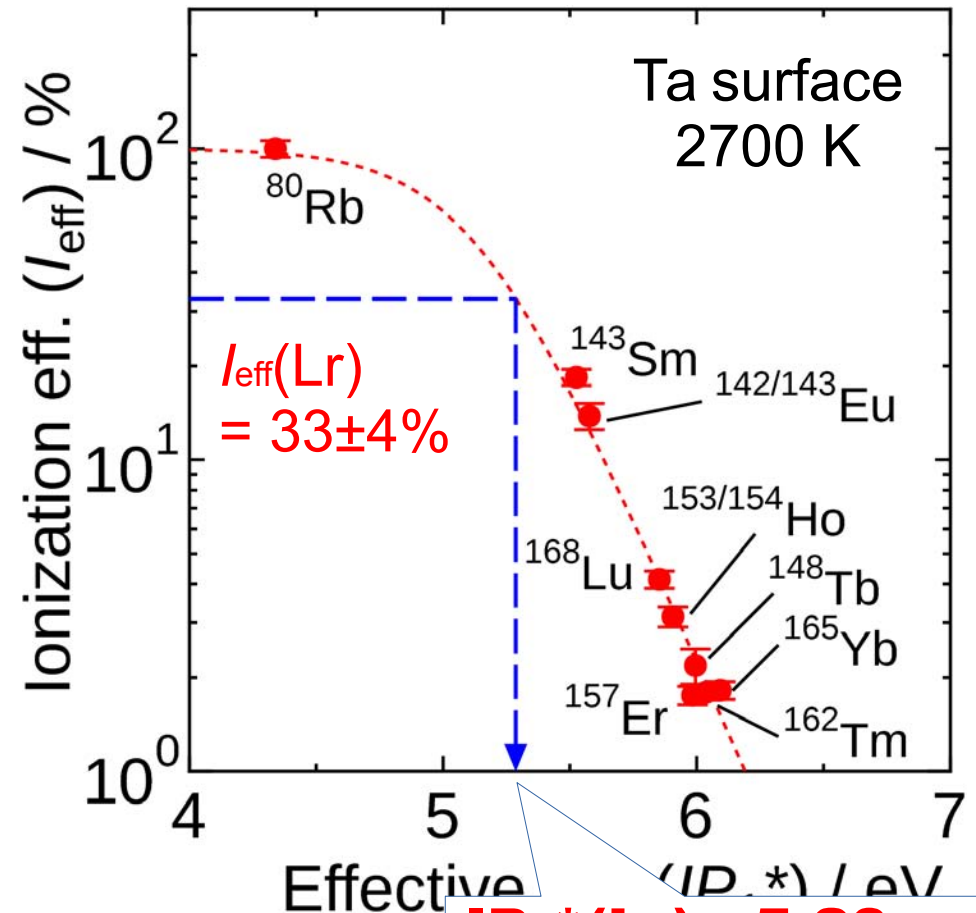
Isotope: ²⁵⁶Lr ($T_{1/2} = 27$ s)
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Ionization temp.: 2700K



$I_{\text{eff}}(\text{Lr}) = 33 \pm 4\%$
in the present system



$\text{IP}_1(\text{Lr}) = 4.96 \pm 0.08$ eV



$\text{IP}_1^*(\text{Lr}) = 5.29$



$\text{IP}_1(\text{Lr}) = 4.96$

IP₁ measurement of heavy actinides

^{257}No ($T_{1/2} = 24.5$ s) : $^{248}\text{Cm}(^{13}\text{C}, 4n)$

^{251}Md ($T_{1/2} = 4.27$ min) : $^{243}\text{Am}(^{12}\text{C}, 4n)$

^{249}Fm ($T_{1/2} = 2.6$ min) : $^{243}\text{Am}(^{11}\text{B}, 5n)$

IP₁ measurement of heavy actinides

²⁵⁷No ($T_{1/2} = 24.5$ s) : ²⁴⁸Cm(¹³C, 4n)

²⁵¹Md ($T_{1/2} = 4.27$ min) : ²⁴³Am (¹²C, 4n)

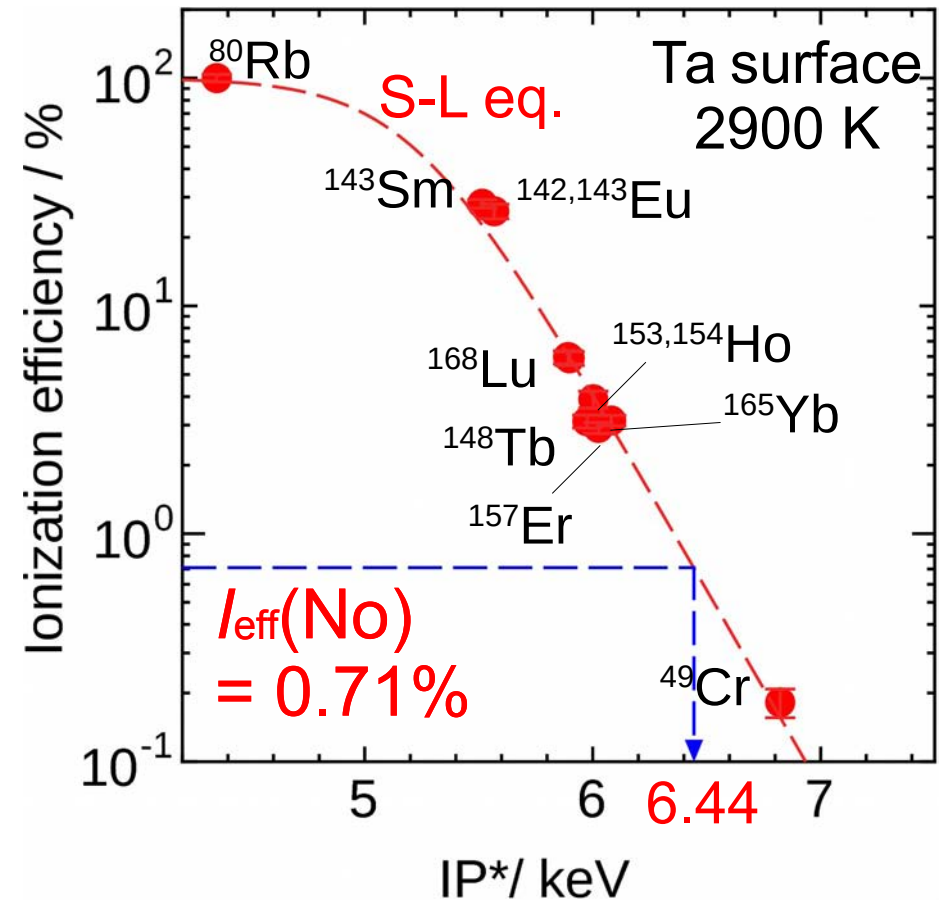
²⁴⁹Fm ($T_{1/2} = 2.6$ min) : ²⁴³Am (¹¹B, 5n)

ex)

$$I_{\text{eff}}(\text{No}) = 0.71 \pm 0.06 \%$$



$$IP_1(\text{No}) = 6.63 \pm 0.08 \text{ eV} \\ \text{(Preliminary)}$$



IP₁ measurement of heavy actinides

²⁵⁷No ($T_{1/2} = 24.5$ s) : ²⁴⁸Cm(¹³C, 4n)
²⁵¹Md ($T_{1/2} = 4.27$ min) : ²⁴³Am (¹²C, 4n)
²⁴⁹Fm ($T_{1/2} = 2.6$ min) : ²⁴³Am (¹¹B, 5n)

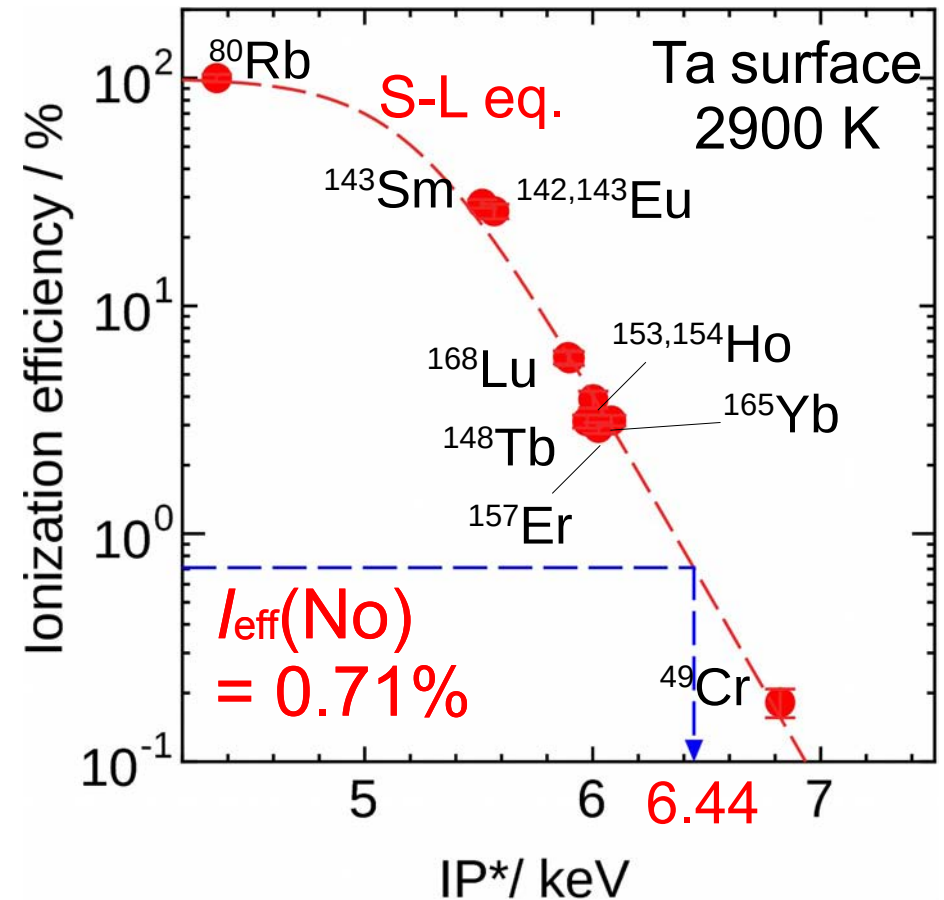
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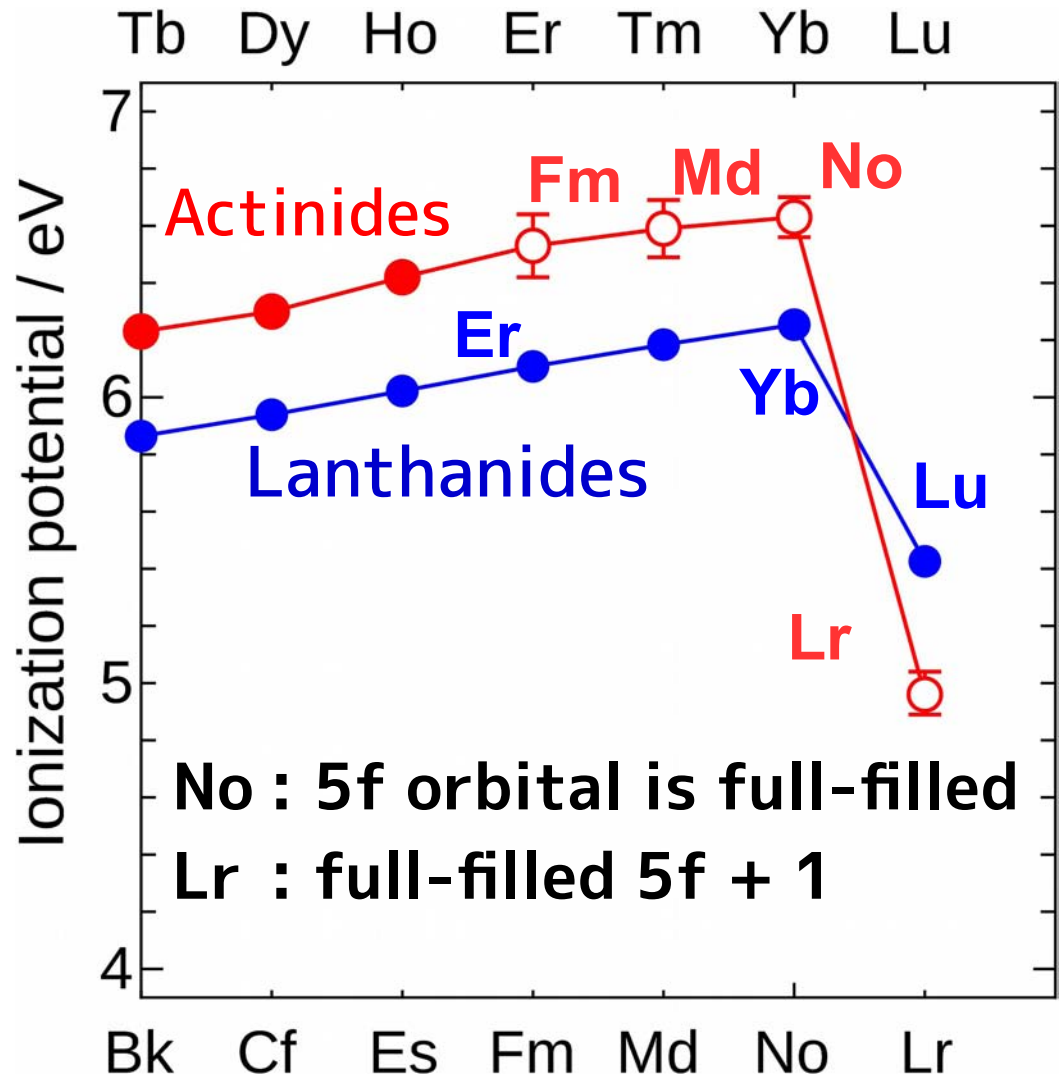
(Preliminary)



	Fm	Md	No
J.Sugar [1]	6.50	6.58	6.65
this work	6.53 ± 0.11	6.59 ± 0.10	6.63 ± 0.08

[1] J. Sugar, J. Chem. Phys. 60 (1974) 4103.

IP₁ measurement of heavy actinides



We successfully measured IP₁ values of Lr ~ Fm using the surface-ionization method.

Lr: IP₁(Lr) = 4.96 ± 0.08 eV

→ **Lr: [Rn]5f¹⁴7s²7p_{1/2}**

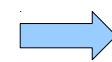
↔ **Lu : [Xe]4f¹⁴6s²5d**

No: IP₁(No) = 6.63 ± 0.08 eV

→ **full-filled 5f orbitals**

Fm → No → Lr

Similar to Er → Yb → Lu



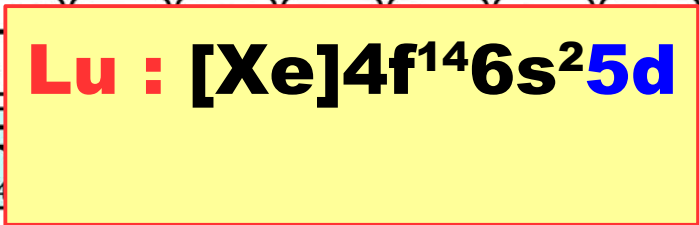
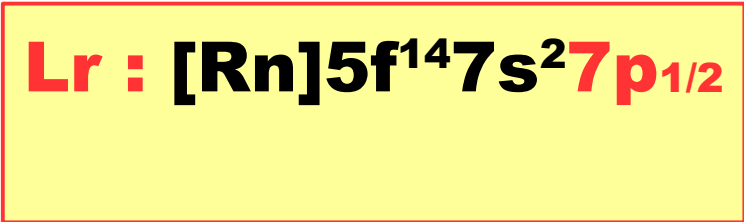
Actinide series would be terminated with Lr.

Present status

Adsorption behavior of Lr on Ta surface

Is Lr a p-element?

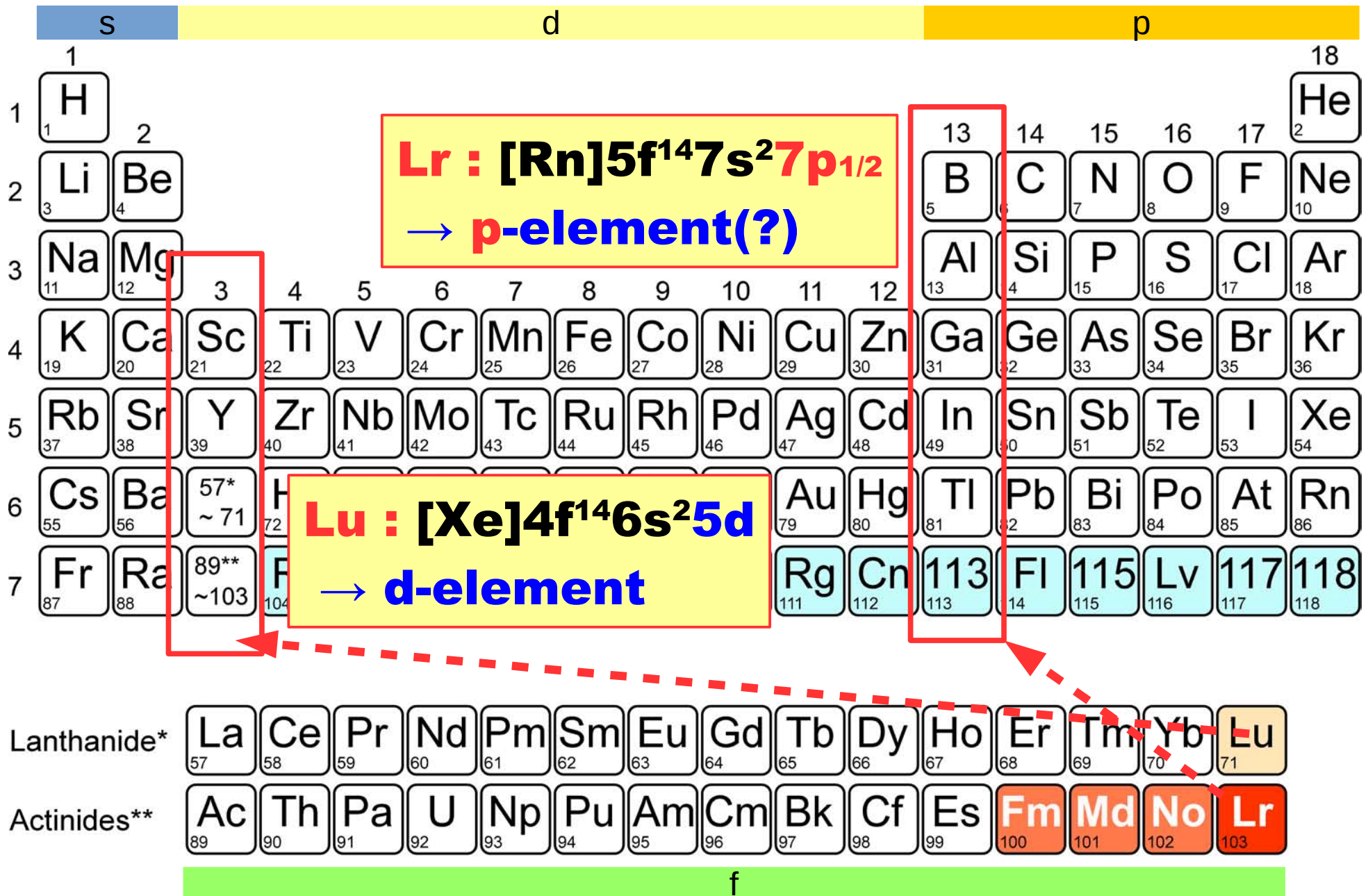
		s		d										p					
1	1	H	2											13	14	15	16	17	18
2	3	Li	4											5	6	7	8	9	10
3	11	Na	12											13	14	15	16	17	18
4	19	K	20	3	4	5	6	7	8	9	10	11	12	31	32	33	34	35	36
5	37	Rb	38	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	49	50	51	52	53	54
6	55	Cs	56	57*	~71							79	80	81	82	83	84	85	86
7	87	Fr	88	89**	~103							111	112	113	114	115	116	117	118



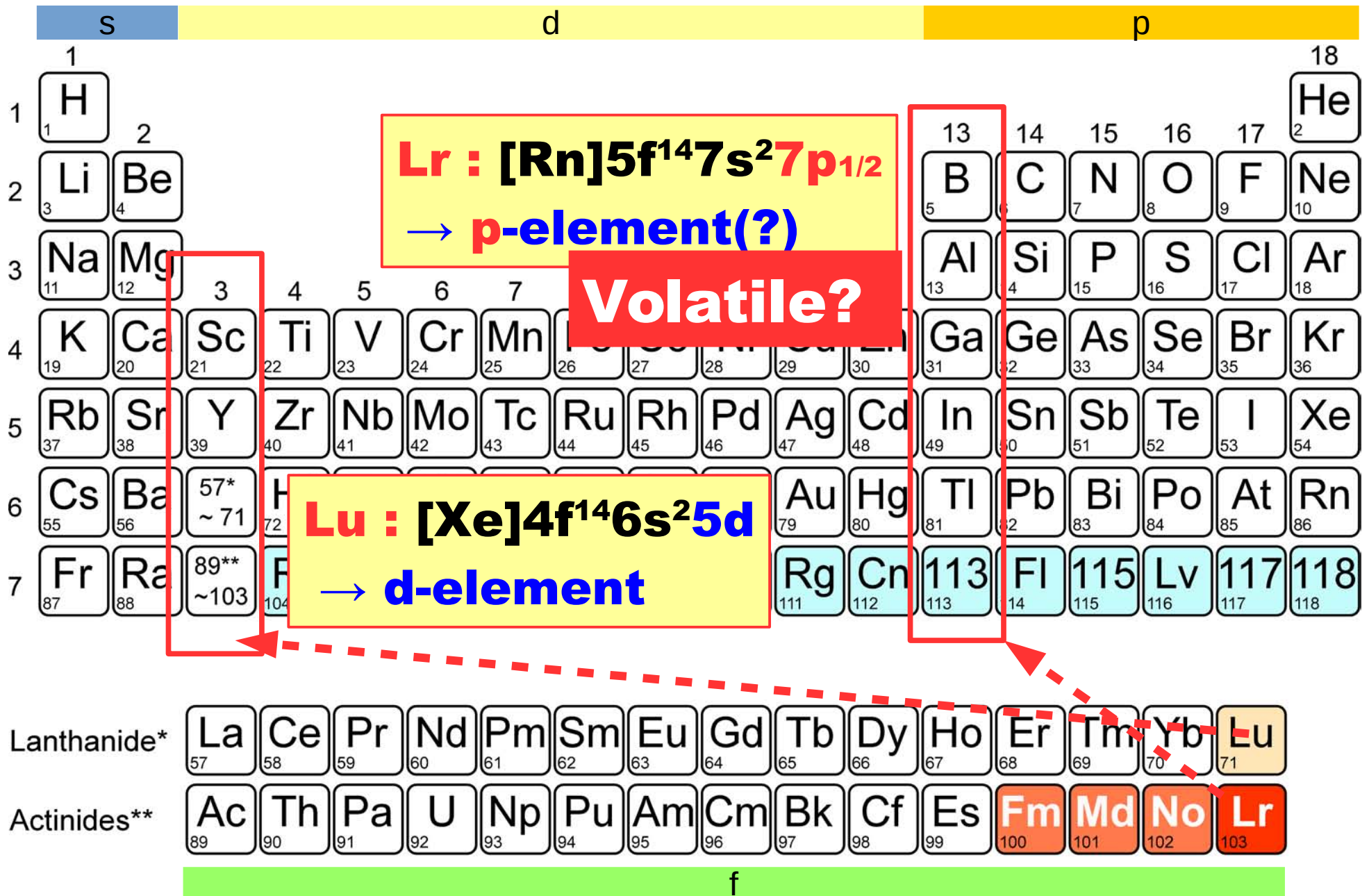
Lanthanide*	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Actinides**	89	90	91	92	93	94	95	96	97	98	99	100	101	102	103
	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

f

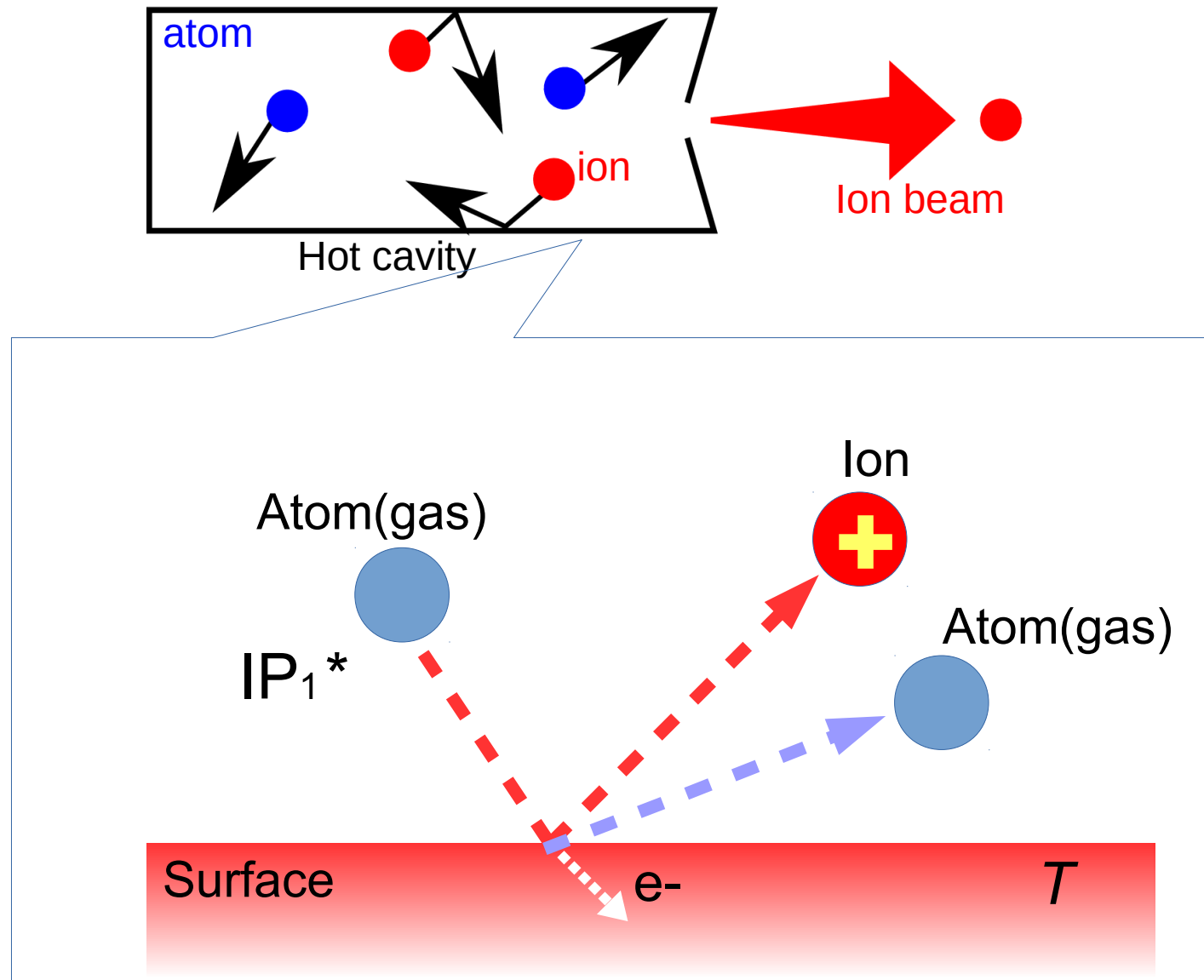
Is Lr a p-element?



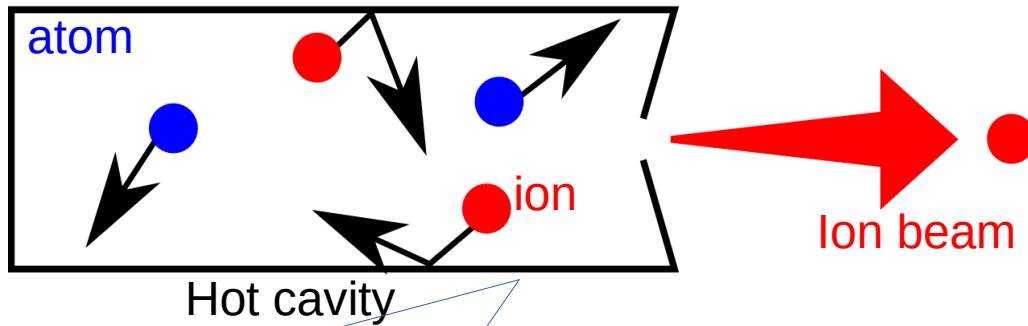
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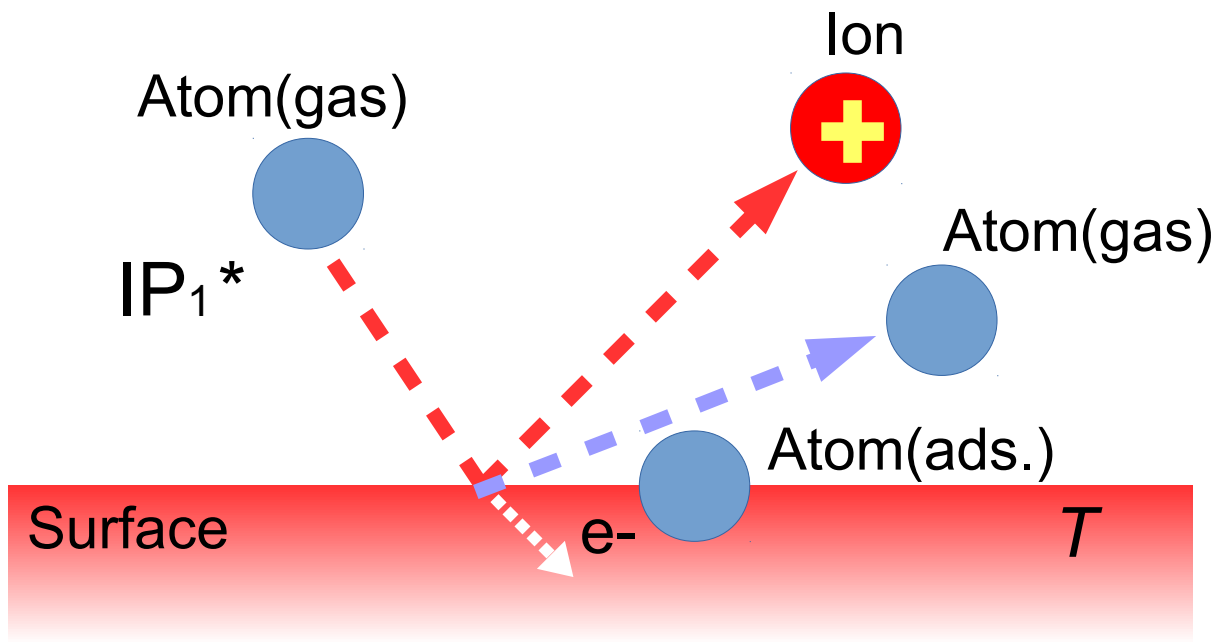
Adsorption on surface of IS



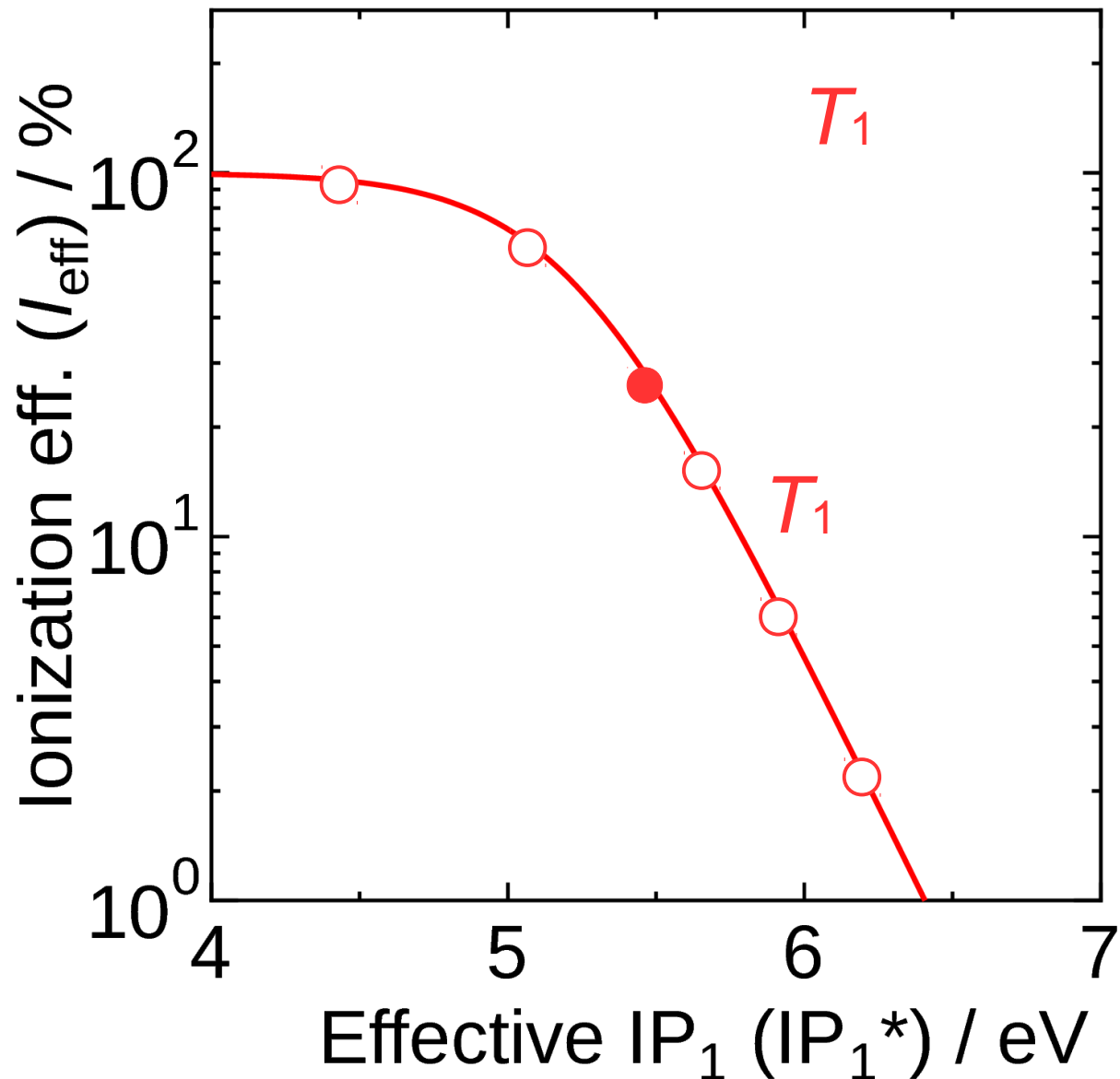
Adsorption on surface of IS



Lower temp.



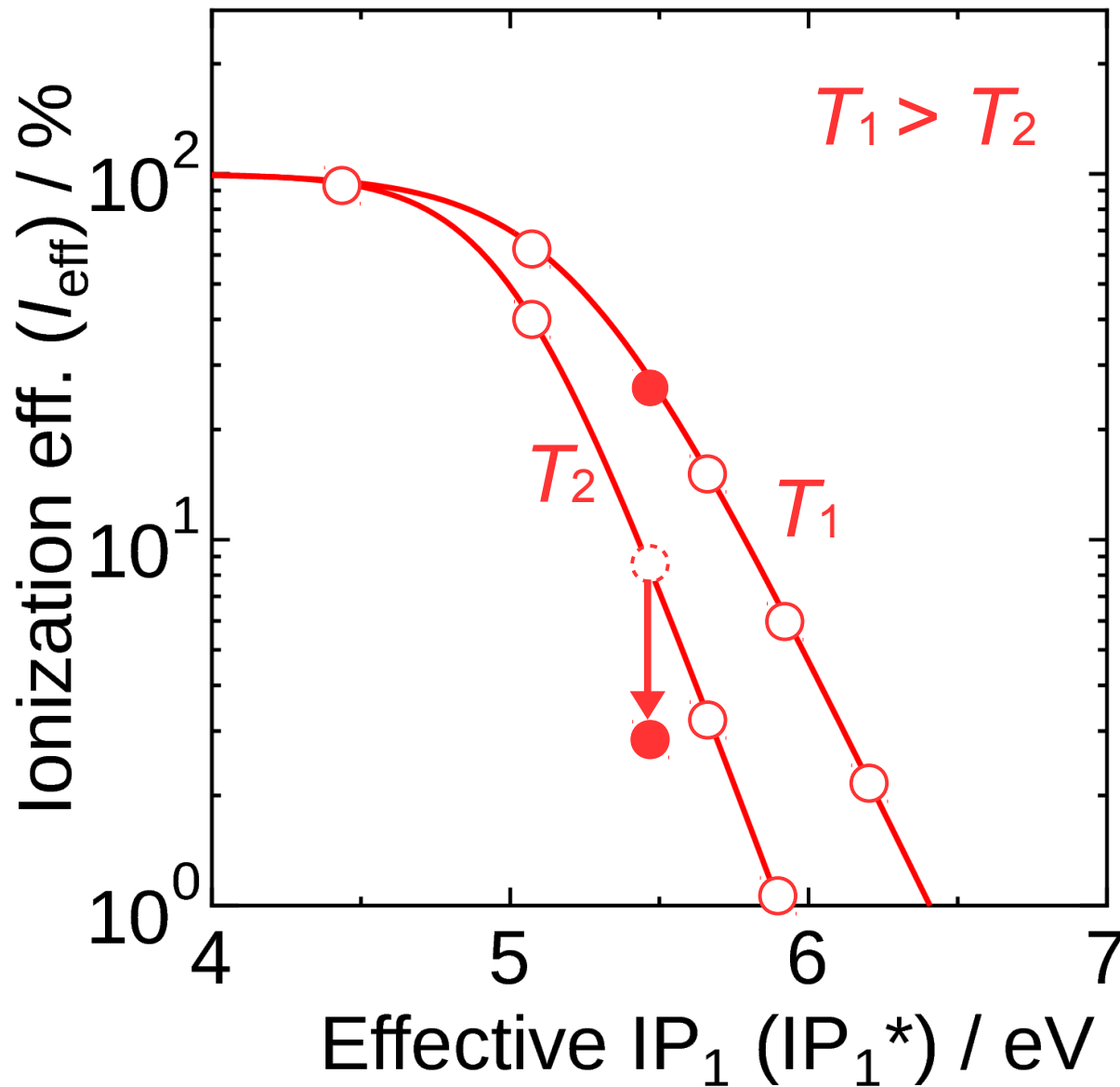
Adsorption on surface of IS



At T_1

$$I_{\text{eff}}(\text{measured}) = I_{\text{eff}}(\text{fitted})$$

Adsorption on surface of IS



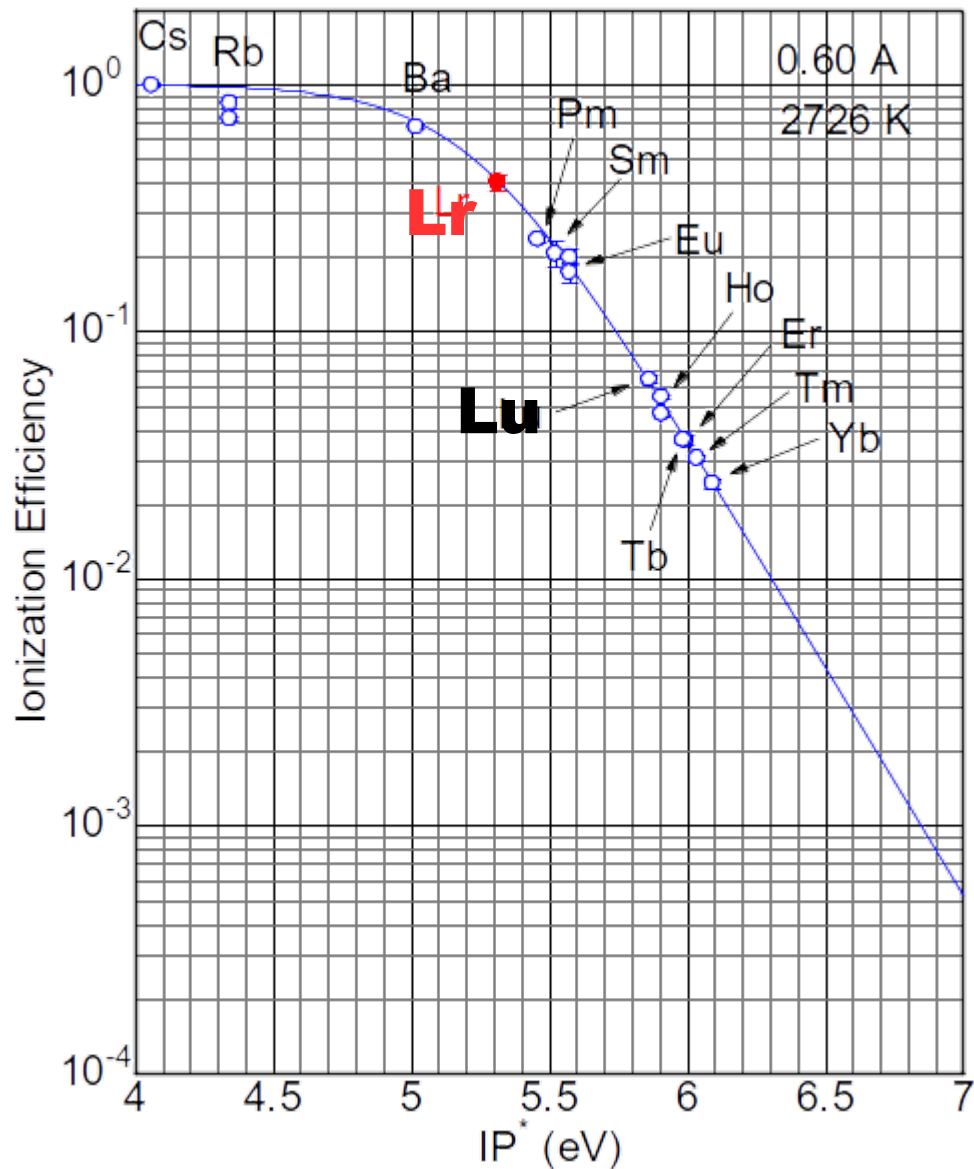
At T_1

$$I_{\text{eff}}(\text{measured}) = I_{\text{eff}}(\text{fitted})$$

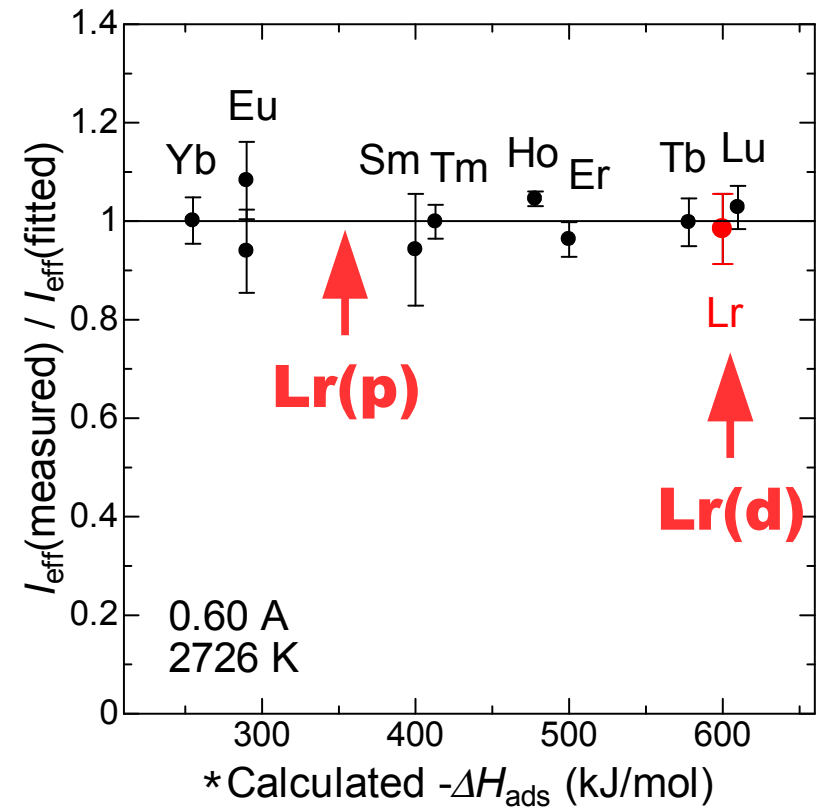
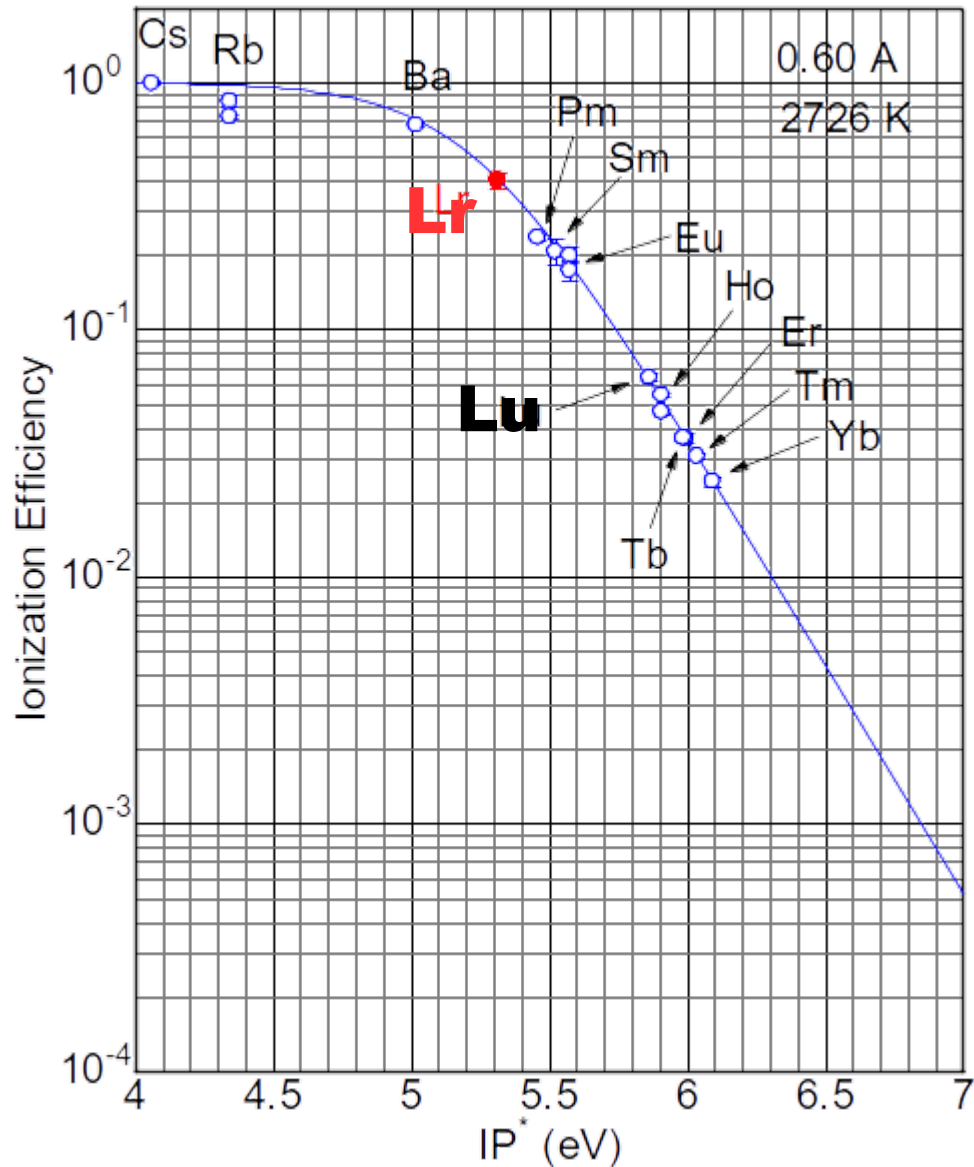
At T_2

$$I_{\text{eff}}(\text{measured}) < I_{\text{eff}}(\text{fitted})$$

Adsorption on surface of IS

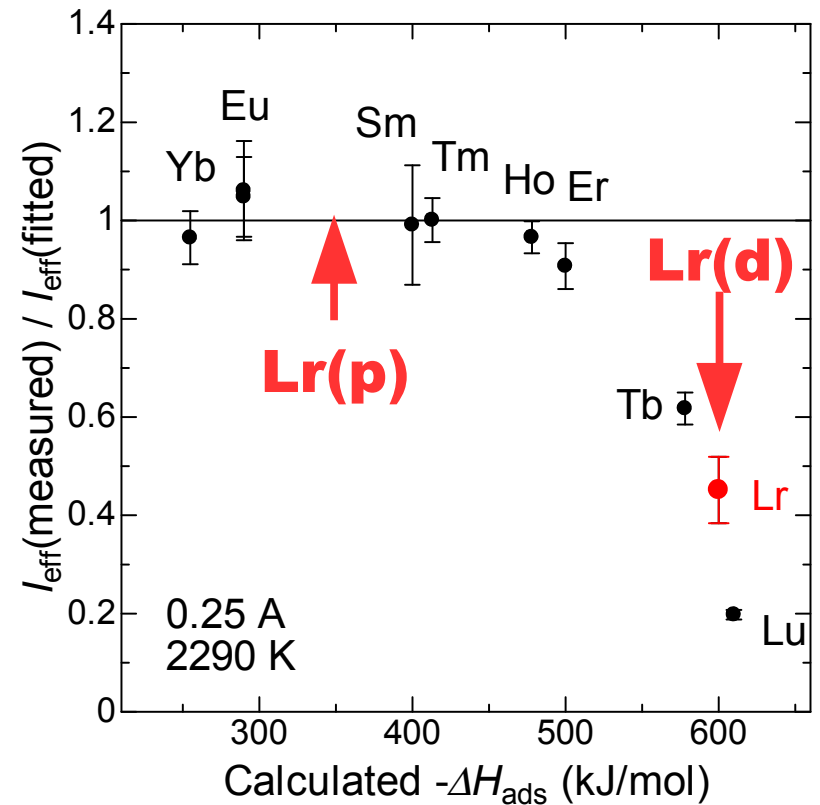
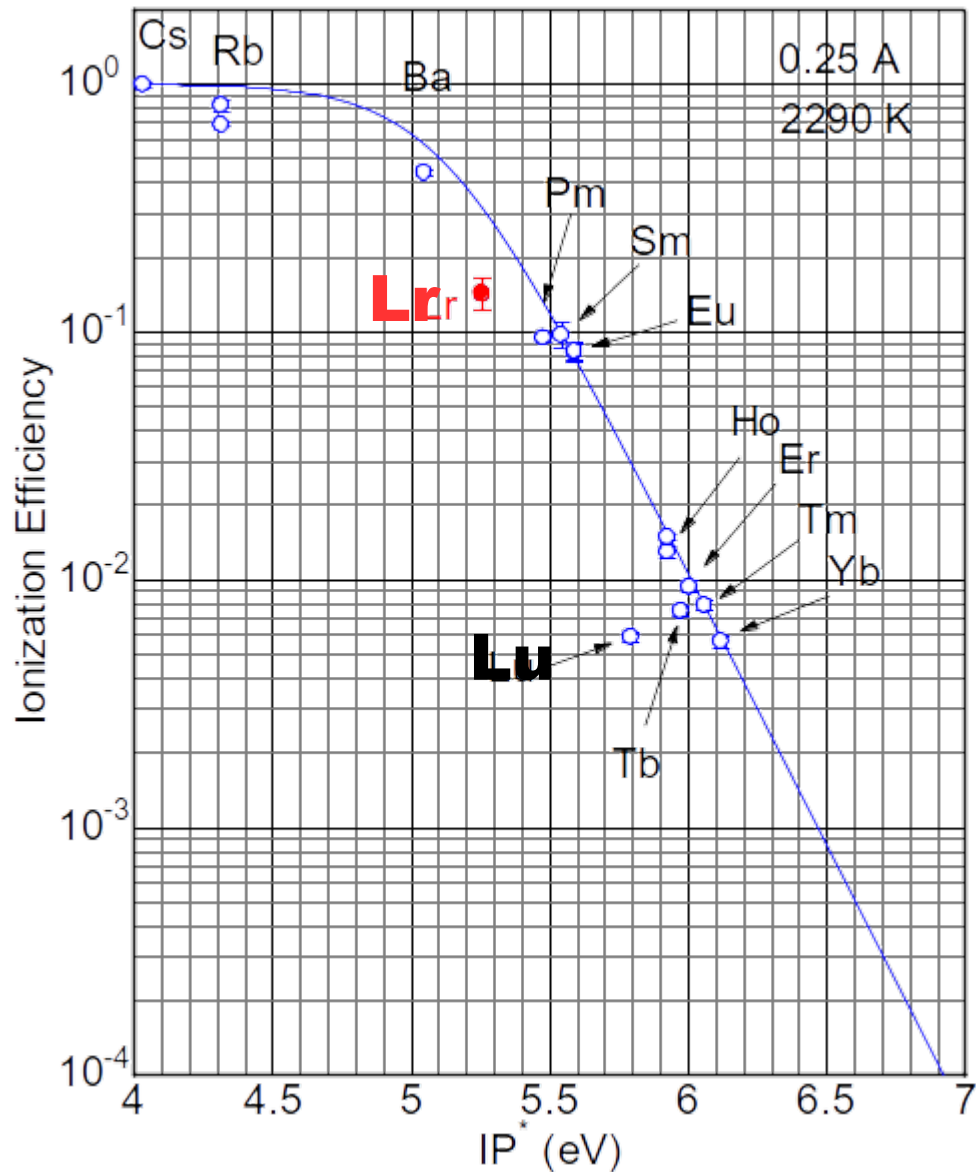


Adsorption on surface of IS

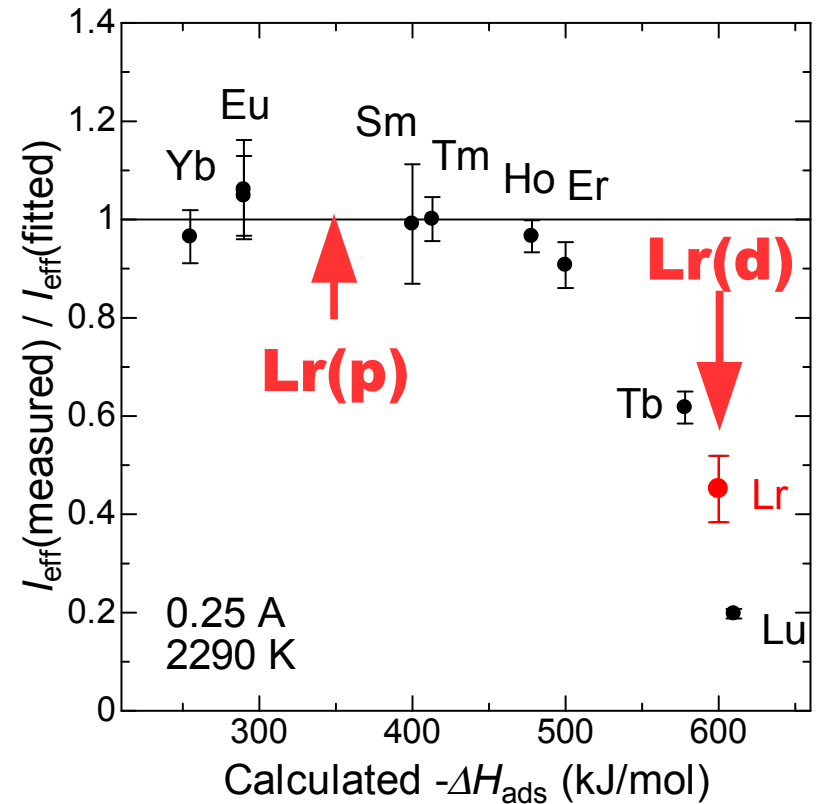
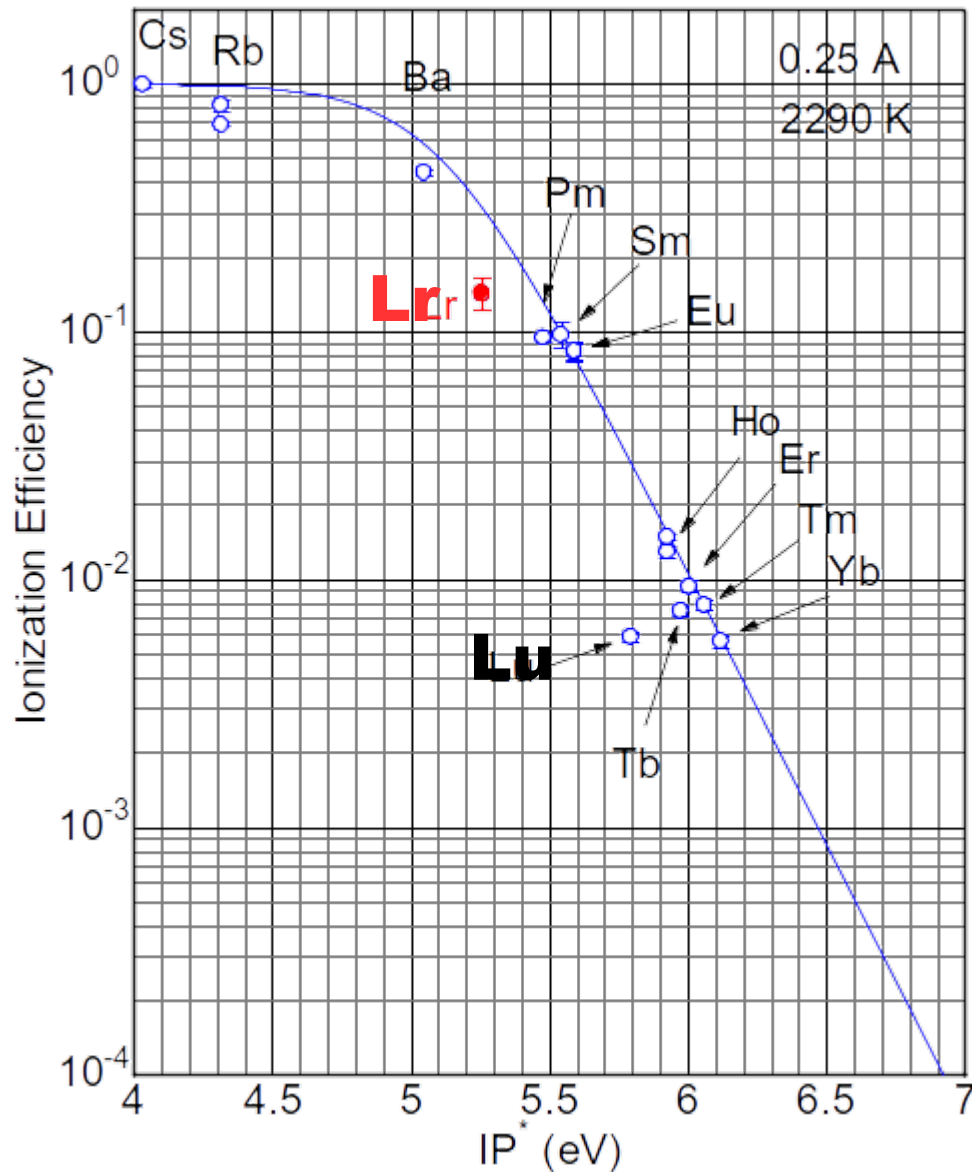


* R. Eichler, private communication

Adsorption on surface of IS



Adsorption on surface of IS



Lr would have a $7p_{1/2}$ electron in the valence orbital like p-element, but its volatility could be similar with that of d-element.