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From the discovery of fission to the synthesis and decay of superheavy nuclei



Ch. Theisen CEA Saclay DRF/IRFU/DPhN Ecole Joliot-Curie 2017



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Since the limits Reaching the limits



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Irfu A lot of room for new isotopes !

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Irfu The discovery of the heaviest elements

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Outline :

- Historical notes : Studies using U decay, reactions with alpha and neutrons
- Fermi neutrons irradiations and evidences for transuranium elements
- The discovery of fission, the liquid drop model
- First transuranium elements
- What is a superheavy nucleus: macroscopic and microscopic views...
- From the chemistry to identification using nuclear properties
- Genetic correlations, separators
- Spectroscopy after alpha decay, interplay with atomic properties
- X-ray identification
- High-K isomers
- Ground states properties : mass measurement and laser spectroscopy
- New facilities
- Naming of the elements

Irfu Subjects not covered in this lecture

- Prompt spectroscopy (including particle spectroscopy after transfer, coulex, ...)
 - Reaction mechanism
 - Fission barrier measurement
 - Shape isomers
 - Search for SHE/SHN in nature
 - Chemistry
 - "Exotic" predictions and phenomena (cluster radioactivity, superdeformed gs, exotic shapes ...)
 - "Exotic" techniques (crystal blocking, lifetime using X-ray fluorescence, ...)
 - Not so much theory
 - ..

Sirfu Historical notes

UNIVERSITE PARIS-SACLAY 1899 Rutherford isolates α and β radioactivities from uranium

1902 Rutherford and Soddy. Emission of $\alpha \rightarrow$ transmutation



1908 Rutherford and Geiger : α = Helium (from thorium emanations)



Ernest Rutherford



Frederick Soddy



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1911 Soddy, Russel : Relation between isotopes after alpha and beta decay



Placement of elements in columns. Chemical similarities with known elements. Rules to change column after alpha and beta decay. A.S. Russell, The Chemical news CVII (1913) 49.



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F. Soddy. Rep. Brit. Ass. Adv. Sci, 83 (1913) 445



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1919 Rutherford Transmutation using α « beam ». α + Nitrogen. First nuclear reaction ! Interpreted as α + Nitrogen \rightarrow p + something Phil. Mag. 37 (1919) 537, 562, 571, 581

1924 Blackett. Visualization of the reaction using a cloud chamber P.M.S. Blackett, Proc. Roy. Soc. A **107**, 349 (1925)



C.T.R. Wilson, Proc. Roy. Soc. A 87, 277 (1912)



 \rightarrow Use of α « beam » to induce nuclear reactions.

Irfu The neutron discovery

UNIVERSITE PARIS-SACLAY 1930. Walther Bothe. Unknown radiations from α + 9Be interpreted as α + $^9Be \rightarrow {}^{13}C^* \rightarrow {}^{13}C$ + γ

1931 F. Joliot and I. Curie. Interpretation as high-energy protons by Compton effect but inconsistent according to Majorana and Rutherford

1932 Chadwick. More sensitive device. Range of protons and impact of the unknown particle on various gases. \rightarrow Existence of a neutral particle « neutron » having the same mass as the proton







James Chadwick

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Sirfu Artificial radioactivity

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Irène and Fréderic Joliot-Curie, 1934 α (²¹⁰Po source) + ²⁷Al \rightarrow (³⁰P)+ n \rightarrow (³⁰Si) Stable

Then with ¹⁰B, ²⁴Mg, ...

 \rightarrow reactions with α

 \rightarrow application of radioisotopes

→ Speculate production of new radioelements using p, d, n C.R. Acad. Sci. 198 (1934) 254



... Drawback of using of α « beam » to induce nuclear reactions: limited to Z~15 due to coulomb repulsion... Not possible to go beyond. Also rather low yield.

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Irfu Fermi : neutron induced reactions

• Work initiated by Orso Mario Corbino

- Neutron produced using Rn alpha source (800 mC) + Be. Rather low neutron production (1000 n/s/mC) but compensated by high crosssection of neutron-induced reaction
- Systematic investigation in Roma of neutron-induced reaction along the periodic table for H to U.

Methodology

- Irradiation ${}^{A}_{Z}X + n \rightarrow {}^{A+1}_{Z}X \xrightarrow{\beta \ decay} {}^{A+1}_{Z+1}Y$
- (chemical separation)
- Detection of radioactivity (β -)
- Using a Geiger-Müller counter
- \rightarrow lifetime and eventually β energy using absorbers

About 30 new isotopes discovered !







Glass tubes with Rn+Be



Geiger-Müller counter



Cylinder irradiated



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Sirfu I Ragazzi di via Panisperna

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Oscar D'Agostino, Emilio Segrè, Edoardo Amaldi, Franco Rasetti, Enrico Fermi (picture taken by Bruno Pontecorvo ?)

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Irfu Ausonium and Hesperium

Possible Production of Elements of Atomic Number Higher than 92

By PROF. E. FERMI, Royal University of Rome

Nature 133 (1934) 898

In this way it appears that we have excluded the possibility that the 13 min.-activity is due to isotopes of uranium (92), palladium (91), thorium (90), actinium (89), radium (88), bismuth (83), lead (82). Its behaviour excludes also ekacæsium (87) and emanation (86).

This negative evidence about the identity of the 13 min.-activity from a large number of heavy elements suggests the possibility that the atomic number of the element may be greater than 92. If it were an element 93, it would be chemically homologous with manganese and rhenium. This hypothesis is supported to some extent also by the observed fact that the 13 min.-activity is carried down by a precipitate of rhenium sulphide insoluble in hydrochloric acid. However, as several elements are easily precipitated in this form, this evidence cannot be considered as very strong.

(Tc was not yet discovered)

²³⁸U + n
$$\rightarrow$$
 ²³⁹U $\xrightarrow{\beta^{-}}$ ²³⁹93 $\xrightarrow{\beta^{-}}$ ²³⁹94

Elements named Ausonium and Hesperium by Franco Rasetti

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Several decay products found with 10s, 40s, 13 and 90 min lifetime. Attempts to prove due to Z=93 using chemical separation.

IA	II A	III B	IV B	VВ	VIB	VII B	l):	VIII		IB	IВ	III A	IV A	V A	VI A	VII A	0
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 0	9 F	10 N
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 CI	1. A
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Ге	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	Э К
37 Rb	38 Sr	39 Y	40 Zr	41 ND	42 Mo	(13)	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 1	5 X
55 Cs	56 Ba	57-71 Ln	/2 Hf	73 Ta	74 W	75 Re	76 Os	17 11	78 Pt	79 Au	80 Hg	81 TI	82 Pb	83 Bi	84 Po	(85)	8 R
(87)	88 Ra	89 Ac	90 Th	91 Pa	92 U	(93)	(94)	(95)	(96)	(97)	(98)	(99)					
		57	58	59	60	(61)	62	63	64	65	66	67	68	69	70	71	
		1.8	Ce	Pr	Nd	63536	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	

Periodic table in the 1920s-1930s following Moseley's work (identification of new elements using X-ray spectroscopy)



Claim for discovery of element 93 by Odolen Koblic, a Czech engineer. Found in pitchblende ores. Chemical solution acidified with nitric acid then thallium nitrate added «Just as expected a vermillion coloured crystalline sediment appeared ». Chemical analysis using hydrogen sulphide. Bohemium (Bo) in honour to fatherland.

> Chemiker-Zeitung 28 (1934) 581 Retracted the same year (Koblic, O. Chem. Obzor. 9 (1934) 146)



Odolen Koblic

Irfu 1938 : Fermi Nobel lecture

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December 10, 1938

- "We concluded that the carriers were one or more elements of atomic number larger than 92; we, in Rome, use to call the elements 93 and 94 Ausenium and Hesperium respectively."
- After the Nobel lecture, Fermi leaves to the US.
- The Roma group was already dispersed → no continuation of the transuranium neutron-induced studies from 1935
 - Rasetti 1935 \rightarrow US \rightarrow Canada
 - Pontecorvo 1936 \rightarrow France then Canada then UK then URSS
 - Segre $1938 \rightarrow US$
 - Amaldi 1939 \rightarrow US Footenote in Fermi's lecture :



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<mark>>2</mark> Irfu	Element 93 confirmed at E more !	Berlin and much					
UNIVERSITE PARIS-SACLAY	1935 : neutron induced reaction repeated by chemists Hahn, Meitner and Strassmann at Kaiser Wilhelm-Institut far Chemie, Berlin (and in other places)						
	Compared to Fermi group, improved clifetime component identified and bette	hemical separation, more or lifetime measurement.					
1. $U + n - \frac{6}{5}$ 2. $U + n - \frac{3}{5}$ 3. $U + n - \frac{3}{5}$	$ \xrightarrow{10''} U \xrightarrow{\beta} \stackrel{2,2'}{93} Eka \operatorname{Re} \xrightarrow{\beta} \stackrel{59'}{94} Eka \operatorname{Os} \xrightarrow{\beta} \\ \xrightarrow{6h} Eka \operatorname{Ir} \xrightarrow{\beta} \stackrel{2,5h}{96} Eka \operatorname{Pt} \xrightarrow{\beta} \stackrel{97}{97} Eka \operatorname{Au}? \\ \xrightarrow{40''} U \xrightarrow{\beta} \stackrel{16'}{93} Eka \operatorname{Re} \xrightarrow{\beta} \stackrel{5,7h}{94} Eka \operatorname{Os} \xrightarrow{\beta} \stackrel{95}{95} Eka \operatorname{Ir}? \\ \xrightarrow{23'} U \xrightarrow{\beta} \stackrel{93}{93} Eka \operatorname{Re}? $ Meitner, Hahn, Strassmann. ZP 106 (1937) 249						
	P. Abelson using the Berkeley Cyclotron	Otto Hahn, Lise Meitner					



found.

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as a neutron source (large flux) \rightarrow no

conclusive results, no alpha decay

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UNIVERSITE PARIS-SACLAY 1938 Irène Curie and Pavel Savitch. New approach: first counting without separation \rightarrow a new β - 3.5 h activity, but chemistry uncertain (looks like La) C.R; Acad. Sci. 206 (1938) 906, 1643

Hahn and Strassmann, activity follows a Ba carrier \rightarrow isotope of Ra (in the same column) ?

Meitner leaves Germany, still close contact With Hahn. Some doubts on the Ra result (need two α emissions).

Hahn and Strassmann. Fractional crystallization (M. Curie method)

 \rightarrow No Ra

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 \rightarrow product is Ba

O. Hahn and F. Strassmann, Naturwiss 27 (1939) 11 (in German).

A result that "contradicts all the

experiences of nuclear physics to date"



Fritz Strassmann





Hahn-Meitner-Strassmann device at Deutsches Museum, **Munich**





UNIVERSITE PARIS-SACLAY Christmas 1938 : Lise Meitner meets his nephew Otto Frisch in Sweeden. During a hike outdoor, they discuss recent results by Hahn and Strassmann, and conceive the fission process.

Estimate energy released by fission ~ 200 MeV using the liquid drop model.

L. Meitner and O. Frisch, Nature 143 (1939) 239



Sission, interpretation

Jan 1939 :

 Frisch discusses with Bohr in Copenhagen "Oh, what idiots we all have been ! Oh but this is wonderful ! That is just as is must be !"*

Frisch reminiscences « What little I remember », 1979

- Frisch first detects the fission fragments from uranium using an ionization chamber → Nature 143 (1939) 276
- Fission also detected by Herbert Anderson et al, US. PR 55 (1939) 511
- Evidences that huge energy production is possible
- Frédéric Joliot detects fission fragment C.R. Acad. Sci 208 (1939) 341 (1939); J. phys. et radium 10 (1939) 159

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Spring 1939 : Theory of fission by Bohr and Wheeler
(PR 56 (1939) 426), Frenkel (PR 55 (1939) 987)
using the liquid drop model
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Dec. 1939 : about 100 papers on fission published !



Yakov Frenkel

Slow neutrons

• 1934, Pontecorvo, Amaldi. Ag irradiation by neutron : more efficient on a

- wood table compared to rock or metal
- Paraffin more efficient
- Water in garden fountain even more efficient !
- \rightarrow neutrons slow-down by H
- \rightarrow neutrons spent more time
- in the nucleus \rightarrow higher cross-section
- E. Fermi et al La Ricerca Scientifica 5 (1934), 282







Irfu The liquid drop model

Early versions by G. Gamow (1929), W. Heisenberg (1933) to account for the mass-defect of the nuclei (Aston curve)



G. Gamow. Proc. Roy. Soc. A 126 (1930) 632 Water drop of α particles with surface tension

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Irfu The liquid drop model



W. Heisenberg, *Considérations théoriques sur la structure du noyau* (in French !), congrès Solvay 1933

Continuation by Carl Friedrich von Weizsäcker (Heisenberg's student).



W. Heizenberg, C.F. von Weizäcker 1935

Irfu The liquid drop model

UNIVERSITE PARIS-SACLAY	The Bethe - Weizsäcker mass formula					
	$BE(A,Z) = a_v A$	Volume → attractive → short interaction range → binding energy ~ constant = saturation				
	- a _c Z²/A ^{1/3}	Coulomb \rightarrow repulsive				
	-a _s A ^{2/3}	Surface : less neighbours → repulsive (re)introduced by von Weizsäcker (1935) Z. Phys. 96 (1935) 431				
	-a _a (N-Z) ² /A	Asymmetry				
	+ δ(A,Z)	Pairing introduced by Bethe and Bacher (1936) Rev. Mod. Phys. 8 (1936) 82 "the bible"				
	Warning : liquid drop is not a phenomenological model, it is based on fir principles although in practice parameters are fitted on known masses 1939 Bohr and Wheeler, Frankel Stability = balance between coulomb and surface terms					

Irfu Deformed liquid drop and fission barrier

Energy of a deformed liquid drop :

$$E_{\rm C}(a) = E_{\rm C}(0) \left(1 - \frac{1}{5}a^2 - \frac{4}{105}a^3 + \dots \right), \qquad a = \sqrt{\frac{5}{4\pi}}\beta_2,$$
$$E_{\rm S}(a) = E_{\rm S}(0) \left(1 + \frac{1}{5}a^2 - \frac{4}{105}a^3 + \dots \right).$$

Change of energy as a function of deformation :

$$\Delta E = E_{\rm S}(a) + E_{\rm C}(a) - E_{\rm S}(0) - E_{\rm C}(0)$$

= $E_{\rm S}(0) \left[\frac{2}{5} (1-x) a^2 - \frac{4}{105} (1+2x) a^3 + \dots \right].$
 $x = \frac{1}{2} \frac{a_{\rm C} Z^2 / A^{1/3}}{a_{\rm S} A^{2/3}} = \frac{a_{\rm C} Z^2}{2 a_{\rm S} A}.$

Liquid drop instable if x>1 \rightarrow Z²/A \gtrsim 48 x = fissility parameter

 ^{238}U + n \rightarrow ^{239}U + excitation energy \rightarrow fission although x = 0,77

Irfu Deformed liquid drop and fission barrier

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Irfu Liquid-drop fission barrier and lifetime

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$$B_f = \frac{98}{15} \frac{(1-x)^3}{(1+2x)^2} Es(0).$$

Penetration through the barrier : Wentzel–Kramers–Brillouin–Jeffreys semi-classical approximation \rightarrow

$$T_{1/2}(s) = \ln 2 \ 10^{-21} \exp(2\pi B_f / \hbar \omega_f)$$

 $\hbar \omega_f$: barrier curvature ~ 0,5 meV

Nucleus	x	B _f LDM	T _{1/2} (s) LDM
²³⁸ U	0.77	7.76	1.6 10 ²¹
²⁴⁰ Pu	0.79	5.8	3.6 10 ¹⁰
²⁵⁵ Fm	0.84	2.45	1.5 10 ⁻⁸
²⁵⁴ No	0.86	1.45	6 10 ⁻¹⁴
²⁵⁶ Rf	0.89	0.85	3 10 ⁻¹⁷
²⁹⁰ FI	0.96	0.04	1.1 10 ⁻²¹

Warning : nuclei assumed spherical in their ground-state. Deformation systematics came later (eg Townes 1949)

Spontaneous fission ?

UNIVERSITE PARIS-SACLAY Predicted by Bohr & Wheeler in their seminal paper

Although nuclei for which the quantity Z^2/A is slightly less than the limiting value (11) are stable with respect to small arbitrary deformations, a larger deformation will give the long range repulsions more advantage over the short range attractions responsible for the surface tension, and it will therefore be possible for the nucleus, when suitably deformed, to divide spontaneously. Particularly important will be

Predicted lifetime ~ 10^{30} s ~ 10^{22} years for 239 U Physical Review 56 (1939) 426

Search for spontaneous fission by chemist W.F. Libby, 1939 (Berkeley) Detection of neutrons \rightarrow Uranium, thorium T1/2 > 10¹⁴ year Phys. Rev. 55 (1939) 1269



Niels Bohr



John Archibald Wheeler (selfie !)

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Irfu Consequences of the liquid drop

- **ie** 1 : heavy nuclei can fission spontaneously
 - 2 : fission releases energy
 - 3 : one can estimate the $Q_{\beta\text{-}},\,Q_{\beta\text{+}},\,Q\alpha$ decay energies
 - 4 : most stable nuclei = Beta line of stability « Green approximation »



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Spontaneous fission by Flerov & Petrzhak

Universite Context = possible use of nuclear energy

- Can be produced using ²³⁵U, but problem = isotopic separation (only 0,7 % ²³⁵U in natural U).
- Work investigated by I. Kurchatov : search for alternate solutions (²³⁸U in particular) using different neutron energies
- Work performed by two young collaborators : Flerov & Petrzhak



G.N Flerov and Konstantin Petrzhak, 1940

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Georgy Nikolayevich Flerov, 1940



Igor Kurchatov, 1933

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Sirfu Multilayer ionization chamber



Multilayer fission ionization chamber: 15 plates area = 1000, 6000 cm2, uranium oxide $\rho \approx 10-20$ mg/cm²

Signal without neutron beam : ~ 6 counts / hour Several cross-checks : vibrations, electronics noise, alpha pilup, gas discharge, several chambers, effect related to U quantity, measurement of the signal, amplitude (about 160 MeV).


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Fission induced by cosmic rays ?

→ test in a Moscow subway station (Dinamo) 50 m underground

Spontaneous Fission of Uranium

With 15 plates ionization chambers adjusted for detection of uranium fission products we observed 6 pulses per hour which we ascribe to spontaneous fission of uranium. A series of control experiments seem to exclude other possible explanations. Energy of pulses and absorption properties coincide with fission products of uranium bombarded by neutrons. No pulses were found with UX and Th. Mean lifetime of uranium follows ten to sixteen or seventeen years.

> Flerov Petrjak

PR 58 (1940) 89

• Shortest nuclear physics paper ever ?

Radium Institute (P), Leningrad, U. S. S. R., June 14, 1940 (by cable).

- Kurchatov not signing the paper
- Which U isotope ? (later identified as ²³⁸U).

Physico Technical Institute (F),

• Lifetime = ?

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More detail in Russian journals

Reminiscences in Petrzhak & Flerov : Soviet. Phys. Uspekhi 4 (1961) 305

• No reaction from the west countries....

<mark>∕</mark>Irfu Idiots ?

UNIVERSITE PARIS-SACLAY Alternative interpretation of Fermi experiments by I.Noddack Angewandte Chemie 37 (1934) 653 (in german)

"It is conceivable, that when heavy nuclei are bombarded by neutrons, these nuclei break up into several larger fragments,



Ida Noddack

which would of course be isotopes of known elements but not neighbours of the irradiated elements."

But comment ignored. Noddack's reputation was not that good in particular since she claimed discovery of Z=43 which could not be verified.

Irfu Fission was already postulated in 1930 !

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Henry A. Barton. Phys. Rev. 15 (1930) 408

« A new regularity in the list of existing nuclei » A paper in a series trying to explain regularities in (e-,p) plots (it was still belived that nuclei we built from electron and protons only). This kind of work lead to evidences for the shell model.

and electrons as the center of the symmetrical cluster. Such a nucleus has not been found to exist in the earth's crust and may hypothetically be regarded as unstable. Suppose there to be a tendency on the part of this nuclear type to break into just two nearly (but not precisely) equal parts. The products of any one such event would be (80+X, 45+Y) and, since the second part is postulated to contain the rest of the nuclear matter, (80-X, 45-Y). Obviously there would thus come into existence always two nuclei symmetrically located X. Y and -X. -Y units respectively from the symmetry center (80, 45). The possible values X, Y would presumably be governed by nuclear forces of cohesion. That is, like a crystal, the nucleus might have particular surfaces of division more probable than others.

Actually Barton postulated fission !!

... and asymmetric fission modes !

(speculation not based on anything, and which does not explain the regularities)

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What was observed by Fermi, Hahn & Irfu Strassmann, Curie and Savitch ${}^{66}_{95}$ Eka Ir $\xrightarrow{\beta}{}^{2,5h}_{96}$ Eka Pt $\xrightarrow{\beta}{}_{97}$ Eka Au? 2. $U + n \longrightarrow {}^{40''}_{92}U \xrightarrow{\beta} {}^{16'}_{93}Eka \operatorname{Re} \xrightarrow{\beta} {}^{5,7h}_{94}Eka \operatorname{Os} \xrightarrow{\beta} {}^{95}_{95}Eka \operatorname{Ir}?$ 3. $U + n \longrightarrow {}^{23'}U \xrightarrow{\beta} {}_{03}Eka Re^{2}$ Correct ! $_{56}Ba \rightarrow _{57}La \rightarrow _{58}Ce \rightarrow _{59}Pr \rightarrow _{60}Nd \rightarrow ...$ \sim_{36} Kr \rightarrow_{37} Rb \rightarrow_{38} Sr \rightarrow_{39} Y \rightarrow_{40} Zr \rightarrow_{41} Nb $\rightarrow \dots$ Experiment repeated 1971 : H. Menke, G. Herrmann. Rad. Acta 16 (1971) 119 At least 22 fission products 66h : ⁹⁹Mo (67h) + ¹³²Te (78hr) 2.5h : ¹³²I (2,26h) Other complicated mixtures e.g. $16 \text{min} = {}^{101}\text{Tc} + {}^{101}\text{Mo} + {}^{131}\text{Sb} + {}^{131}\text{Te} + {}^{130}\text{Sb}$ (18min)

3.5 h Curie & Savitch activity : mixture of Y and La isotopes Herrmann, radioch. Acta 3 (1964) 164.

Sequanium Z=93

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Horia Hulubei and Yvette Cauchois.

Search for element 93 in natural samples.

Analysis of minerals betafite from Madagascar, tantalite from France. Chemical analysis + Xray spectroscopy. C.R. Acad. Sci 209 (1939) 476





Irfu Discovery of elements 93, 94

UNIVERSITÉ PARIS-SACLAY 1930's : first electrostatic accelerator by John Douglas Cockroft and Ernest Walton, cyclotron by Ernest Lawrence

Very fast development of cyclotrons in the US then in other countries: Russia (1934), UK (1935); France(1937), Japan (1937), Denmark (1938); Sweeden (1938), ...

1933 production of neutrons using a 27 inch cyclotron at Berkeley : M. S. Livingston, M. C. Henderson, and . E.O. Lawrence. d (1.3 MeV, 10^{-8} A) + 9 Be \rightarrow 10 Be+n ~ 500 000 n/s. PR 44 (1933) 782



Livingston and Lawrence, 27" cyclotron

Septunium

UNIVERSITE PARIS-SACLAY 1939 : Edwin Mc Millan and Emilio Segré. Berkeley Cyclotron. Neutron from d(8MeV) + ⁸Be reaction.

23-min activity from ²³⁹U isotope.

Observe a 2.3-day activity. Daughter of ^{239}U ? Chemistry \rightarrow rare-earth. PR 55 (1939) 510, 1104

1940 : McMillan and Alberson. Experiments in Berkeley and Washington.

2.3 day activity is not a rare-earth, not homolog to Re. properties similar to U !

Second « rare-earth » group starting from U ? 2.3-day activity is the daughter of the 23-min U activity \rightarrow proof 2.3-day activity corresponds to ²³⁹93; low energy beta particles \rightarrow Unsuccessful search for ²³⁹94

PR 57 (1940) 1185



Edwin McMillan 1940



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Plutonium

Search for element 94 starting from 1940.

McMillan : d(16 MeV)+²³⁸U, continuation by Seaborg, Kennedy, Wahl. New activity 2 ~ days (^{238, 236 or 235}93).

Observation of daughter α activity (proportional counter) with lifetime ~ 50 years \rightarrow $^{238}94$ (modern value = 87,7 years).

Not a formal proof however but letter sent to PR on January 28th, 1941.

Continuation to identify chemically the alpha emitter

 \rightarrow product has chemical properties similar to U, but different to Os Letter sent to PR in March 7th 1941

In parallel continuation of the Mc Millan and Segré work using neutrons

Alpha activity (ionization chamber) of the 239 93 daughter \rightarrow 30000 years (modern value = 24110 years) Letter sent to PR May 29th, 1941

Voluntary restrictions on publications of papers on fission and transuranium elements: potential application for energy production.

(explains why nobody reacted to the discovery of spontaneous fission) CEA DRF Irfu





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Irfu Chemical identification : what was wrong ?

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IA	II A	III B	IV B	VВ	VIB	VII B	Ú.	VIII		IB	IIВ	III A	IV A	VA	VI A	VIIA	0
1 H																	2 He
3 Li	4 Be											5 B	8 C	7 N	8 0	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 CI	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	38 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	(13)	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 1	54 Xe
55 Cs	56 Ba	57-71 Ln	/2 Hf	73 Ta	74 W	75 Re	76 Os	11 It	78 Pt	79 Au	80 Hg	81 TI	82 Pto	83 Bi	84 Po	(85)	86 Rri
(87)	88 Ra	89 Ac	90 Th	91 Pa	92 U	(93)	(94)	(95)	(96)	(97)	(98)	(99)					
		57	58	59	60	(61)	62	63	64	65	66	67	68	69	70	71	
		1.a	Ce	Pr	Nd	1000	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	

Periodic table ~1930 : Z=93 same column as Mn, Tc, Re

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Sirfu The actinide serie



Actinide concept : Glen Seaborg ~ 1944 Table from G. Seaborg, Science 104 (1946) 379

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A.S. Russell, The Chemical news CVII (1913) 49.



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Irfu Element discoveries and errors

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Discovery of new elements : an history full of errors (and fakes)

458. Gerhard Krüss und L. F. Nilson: Studien über die Componenten der Absorptionsspectra erzeugenden seltenen Erden.

(Vorgetragen in der Sitzung vom 25. April von Hrn. Gerhard Krüss.)

E.

Im Anschluss an eine Untersuchung über das Aequivalent und Atomgewicht des Thoriums¹) untersuchten wir auch die anderen neben der Thorerde in den Thoriter von Brevig und Arendal vorkommenden seltenen Erden. Die Nitrate derselben lieferten sehr schöne Absorptionsspectren, welche mehrere, den Didym-, Samarium, Erbin-, Thulium-Verbindungen, sowie jenen der Soret'schen Erde X oder den Holmiumverbindungen eigene Linien aufwiesen.

Eine genauere Messung der Absorptionsspectren der Thoriterden führte nun zu der auffälligen Beobachtung, dass in diesem Falle nur ein Theil einiger den seltenen Erden eigenen Absorptionsstreifen sicht-

Berichte der Deutschen Chelischen Gesellschaft zu Berlin 20 (1887) 2134

Claim for the discovery of 23 lanthanide elements, all wrong

u		IV	'e
PA	RI	S-	SA

. . .

Date	Element	Discoverer	Reference	Page
1919	Asteroid elements Crustaterrium, Primordial matter Terrium Chondrium, Pallasium Siderium Cosmium	P. N. Chirvinsky	Chirvinskii, P. N. Bull. Inst. Polytechn. Don 1919 , 7(Sect. 2), 94.	411
919	"Helium system" "Hydrogen" system	W. D. Harkins	Harkins,W. D. Science 1919 , 50, 577.	445
921	Emilium	P. Loisel	Loisel, P. Compt. Rend. Chim. 1921, 173, 1098.	284
922	Hibernium	J. Joly	Joly, J. Proc. Roy Soc. A 1922 , 102, 682.	270
923	Oceanium	A. Scott	Scott, A. J. Chem. Soc. 1923, 38, 311.	116
1925	Neutronium Neuton Neutronon	A. von Antropoff	von Antropoff, A., <i>Z. angew.</i> <i>Chem.</i> 1925 , <i>38</i> , 971.	444
1933	Element Z = zero	W. D. Harkins	Harkins, W. D. Nature 1933 , 131, 23.	445
1925	Masurium	W. Noddack; I. Tacke; O. Berg	Zingales, R. "From masurium to trinacrium: The troubled story of element 43," <i>J. Chem.</i> <i>Educ</i> , 2005 , <i>82</i> , 221–27.	310
1925	Pragium	G. Druce	Karpenko,V. <i>Ambix</i> 1980 , <i>27</i> , 77; Ref. 44a.	250
925	Dvi-manganese	Dolejšek, J.; Heyrovský, J.	Dolejšek, J.; Heyrovský, J. <i>Nature</i> 1925 , <i>116</i> , 782.	250
926	Illinium	B S. Hopkins, et al.	Hopkins, B S. <i>Nature</i> 1926 , <i>117</i> , 792	296
927	Florentium	L. Rolla, et al.	Rolla, L. Nature, 1927, 119, 637	296
928	Hypon	W. S. Andrews	Andrews, W. S. The Scientific Monthly 1928 , 27(6), 535.	416
1930	Alkalinium	F. H. Loring	Loring F. H. Chem. News J. Ind. Sci. 1930, 140, 178.	253
1931	Virginium (verium)	F. Allison	Allison, F.; Murphy, E. J.; Bishop, E. R.; Sommer, A. L. <i>Phys. Rev.</i> , 1931 , <i>37</i> , 1178.	323
1931	Element 108	R. Swinne	Swinne, R. Wiss. Veroffentlich. Siemens-Konzern 1931 , 10(No. 4), 137.	326
1932	Adyarium Meta-Elements	Jinarajadasa, C.; C. W. Leadbeater	Jinarajadasa, C.; Leadbeater, C. W. <i>Theosophist</i> 1932 , <i>XII</i> , 361.	439

Date	Element	Discoverer	Reference	Page
1932	Alabamine (alabamium, eline)	F. Allison	Allison, F. et al. J. Am. Chem. Soc. 1932 , 54, 613.	328
1933	Néo-actinium Néo-radium Néo-elements	A. Debierne	Debierne, A. Compt. Rend. Chim. 1933 , 196, 770.	151
1934	Ausonium Hesperium	E. Fermi and co-workers	Fermi, E.; Rasetti, F.; D'Agostino, O. <i>Ricerca</i> <i>Scientifica</i> 1934 , 6(1), 9.	316
1934	Bohemium	O. Koblic	Koblic, O. <i>Chem. Obzor</i> 1934 , 9, 129.	327
1937	Eka-iodine Th-F; Gourium Dakin (Dacinum), Dekhine	R. De	De, R. <i>Separate</i> (Bani Press, Dacca) 1937 , 18.	338
1937	Moldavium	H. Hulubei	Hulubei, H. <i>Compt. Rend.</i> <i>Chim.</i> 1937 , <i>205</i> , 854.	323
1938	Sequanium	H. Hulubei; Y. Cauchois	Hulubei, H.; Cauchois, Y. <i>Compt. Rend. Chim.</i> 1938 , 207, 333.	320
1939	Dor	H. Hulubei; Y. Cauchois	Hulubei, H. Bull. Soc. Roum. Phys. 1944 , 45, no. 82, 3; Hulubei, H. Bull. Acad. Roum. 1945 , 27, no. 3, 124.	331
1940	Helvetium	W. Minder	Minder, W. <i>Helv. Phys. Acta</i> 1940, 13, 144.	340
1942	Anglo-helvetium	W. Minder, A. Leigh-Smith	Minder, W.; Leigh-Smith, A. <i>Nature</i> 1942 , <i>150</i> , 767.	342
1963	Sulfénium	M. Duchaine	Duchaine, M. P. J. <i>French</i> <i>Demande</i> (May 4, 1973) 4 pp., CODEN: FRXXBL FR 2149300.	88
1972	T. W. Kow	Zunzenium	Kow, T. W., J. Chem. Educ. 1972 , 49, 59.	392
1997	Quebecium	P. Demers	Demers, P. Le Nouveau Système des Elements: Le Système du Quebecium; Presses universitaires: Montreal, Canada, 1997.	225
2004	Hawkingium	Anastasovski, P. K.	Anastasovski, P. K. AIP Conference Proceedings 2004, 699 (Space Technology and Applications International Forum—STAIF 2004), 1230.	393

 V. Karpenko. «The discovery of supposed new elements: two centuries of errors». Ambix 27 (1980) 77

 Fontani, Costa and Orna «The Lost Elements: The Periodic Table's Shadow Side» Oxford University Press, 2014

Hundreds of wrong or fake discoveries listed !

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- - -

Siscovery of isotopes



57



UniversiteZ=96 Cm : Seaborg 1944 (60" cyclotron) 239 Pu(α,n) $^{\alpha}_{150d}$ PuAECD-2182 report, Chem. Eng. News 23 (1945) 2190

 $\begin{array}{l} \hbox{$Z$=$95 Am: Seaborg 1944 (60'' cyclotron)$}\\ {}^{238}U(\alpha,n)^{241}Pu ~~ \overbrace{10}^{\beta^{-}} {}^{241}Am$\\ \hbox{$AECD$-$2185 report, Chem. Eng. News 23 (1945) 2190$} \end{array}$

Z=97 Bk : Thompson 1949 (60'' cyclotron) $^{241}Am(\alpha,2n)^{243}Bk \xrightarrow{EC}{\rightarrow} ^{234}Cm$ UCRL-669 report, PR 77 (1950) 838

Z=98 Cf: Thompson 1950 (60" cyclotron) $^{242}Cm(\alpha,n)^{245}Cf \xrightarrow{\alpha}{_{44m}} ^{241}Cm$ UCRL-790 report PR 87 (1950) 298, 102 (1956) 747 (mass assignment was wrong in the 1950 paper)









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Irfu Einsteinium (Z=99) and Fermium (Z=100)





First thermonuclear explosion « Mike » November 1rst 1952, Eniwetok Atoll ~10 Mtons

Explosion debris collected by a plane transferred to Los Alamos.

Results obviously classified.

Some new alpha-rays.

Albert Ghiorso, Berkekey obtains some samples. \rightarrow Discovery ²⁵³Es and ²⁵⁵Fm

In total 15 new isotopes discovered : ^{244,245,246}Pu, ²⁴⁶Am, ^{246,247,248}Cm, ²⁴⁹Bk, ^{249,252,253,254}Cf, ^{253,255}Es, ²⁵⁵Fm



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	29	230	231	232	233	234	235	236	237																



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Mike results classified

- \rightarrow no publication of Es, Fm discovery possible
- \rightarrow « soft » synthesis using ²³⁸U(¹⁴N,6n)²⁴⁶Es
- \rightarrow ^{239}Pu ^{252}Cf neutron captures in a material testing reactor

Thompson et al PR 93 (1954) 908, Harvey, et al PR 93 (1954) 1129

Reactions of U²³⁸ with Cyclotron-Produced Nitrogen Ions*

ALBERT GHIORSO, G. BERNARD ROSSI, BERNARD G. HARVEY, AND STANLEY G. THOMPSON Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California (Received November 25, 1953)

T HE acceleration of $N^{14}(+6)$ ions with the Berkeley Crocker Laboratory 60-inch cyclotron¹ has made it possible to study nuclear reactions of these ions with U²³⁸.

The following transmutation products have been observed: 99²⁴⁷(?), 99²⁴⁶, Cf²⁴⁴, Cf²⁴⁶, Cf²⁴⁷(?), Cf²⁴⁸, Bk²⁴³, and other berkelium isotopes not yet identified. The identification of the elements

Ghiroso et al, PR 93 (1954) 257

New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100

A. GHIORSO, S. G. THOMPSON, G. H. HIGGINS, AND G. T. SEABORG, Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California

M. H. STUDIER, P. R. FIELDS, S. M. FRIED, H. DIAMOND, J. F. MECH, G. L. PYLE, J. R. HUIZENGA, A. HIRSCH, AND W. M. MANNING, Argonne National Laboratory, Lemont, Illinois

AND

C. I. BROWNE, H. L. SMITH, AND R. W. SPENCE, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (Received June 20, 1955)

THIS communication is a description of the results of experiments performed in December, 1952 and the following months at the University of California Radiation Laboratory (UCRL), Argonne National Laboratory (ANL), and Los Alamos Scientific Laboratory (LASL), which represent the discovery of the elements with the atomic numbers 99 and 100.

The source of the material which was used for the first chemical identification of these elements was the Los Alamos Scientific Laboratory which provided uranium which had been subjected to a very high instantaneous neutron flux in the "Mike" thermonuclear explosion (November, 1952). Initial investigations at

Ghiroso et al, PR 99 (1955) 1048

⁴ There is unpublished information relevant to element 99 at the University of California, Argonne National Laboratory, and Los Alamos Scientific Laboratory. Until this information is published the question of the first preparation should not be prejudged on the basis of this paper.



Plowshare program in the US on peaceful uses of nuclear explosion (1958-1975)

- 1961-1973 : 27 tests
- Mainly excavation techniques, and neutron flux studies (including ~10 tests for heavy element production).
- e.g. Hutch event June 1969 neutron flux 4,5 10²⁵ neutron/cm²/s
- Heaviest nucleus observed = ²⁵⁷Fm





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Irfu Heavy elements and the r-process

Related questions

Production of super-heavy in nature; r-process : Supernovae explosion

Why nothing heavier than ²⁵⁷Fm in thermonuclear

Explosions ?

Need very neutron rich Fm nuclei to reach Beta-decaying nuclei (because Z=100 deformed magic shell gap). But ²⁵⁶⁻²⁵⁸Fm predicted too short lived.

Petermann et al « Have superheavy elements been produced in nature? » EPJA 48 (2012) 122



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Irfu Heavy elements and the r-process



Irfu By-passing the Fm gap...



Search for SHE in nature

ersite A vast program with great hopes (and great fakes)

See e.g. Ter-Akopian and Dimitriev NPA 944 (2015) 177 Korschineka and Kutschera NPA 944 (2015) 190 And references therein

Irfu The limits of the periodic table

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Oveview in « Superheavy elements and the upper limit of the periodic table: early speculations ». H. Kragh. EPJH 38 (2013) 411

- 19th century chemistry \rightarrow no limitation
- Bohr-Sommerfeld atomic physics ca 1920. Electron orbits ~ nuclear size → Z ≤ 137.
- Swinne 1926, atomic physics. Possible existence of « transuranic » long lived elements Z=98-102 then Z=108-110.
- Minimum-time hypothesis « chronon ». Minimum period of revolution. Flind and Richardson 1928 \rightarrow Z < 97
- Cosmic speculations. Long-lived elements descendants of early radioactive state of the universe (Rutherford 1923, Kolhöster 1924, Nernst 1928) → idea that one can find transuranium elements on earth
- Jean 1926 Stellar matter. Center of the stars : elements Z~95.
- Lemaitre 1931. Early universe = giant atom of $\sim 10^{54}$ g
- G. Fournier. Geometric lattice model of the nucleus. Maximum size of the nucleus. Z=136, A=360. C.R. Acad. Sci. 203 (1936) 1495

Irfu Limit of stability : positron emission

Nuclei for Z larger than 173 become unstable against positron emission.

This is because the most deeply bound electrons from the $1s_{1/2}$ shell reach an energy of -511 keV



Fig. 2. Binding energies of electronic states in atoms as function of nuclear charge Z. At $Z_c = 173$ the 1s-state dives into the negative energy continuum.

See eg W. Pieper, W. Greiner Z. Phys. A 218 (1968) 327 J. Reinhardt et al, Z. Phys. A 303 (1981) 173

Fission vs liquid drop model

	Nucleus	x	B _f LDM	T _{1/2} (s) LDM	T _{1/2} (s) exp
	²³⁸ U	0.77	7.76	1.6 10 ²¹	0.6 10 ²³
	²⁴⁰ Pu	0.79	5.8	3.6 10 ¹⁰	3.6 10 ¹⁸
	²⁵⁵ Fm	0.84	2.45	1.5 10 ⁻⁸	3.2 10 ¹¹
	²⁵⁴ No	0.86	1.45	6 10 ⁻¹⁴	2.9 10 ⁴
	²⁵⁶ Rf	0.89	0.85	3 10 ⁻¹⁷	6.2 10 ⁻³
	²⁹⁰ FI	0.96	0.04	1.1 10 ⁻²¹	

Swiatecki 1955 : correcting the liquid drop-model for shell structure may improve the description of spontaneous fission half-lives PR 100 (1955) 937



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Wheeler phenomenological approach. « Superheavy » nuclei

After the discovery of the first transuranium elements (up to Fm), the limits of nuclear matter were not at the heart of discussion.

In 1955, John Wheeler coined the term « superheavy » during the (famous) Geneva International conference on the peaceful uses of atomic energy



Estimates based mostly on the liquid drop model. No shell effects included, although the Nilsson Model was known and used to discuss fission barriers (by John Wheeler itself). Calculations using both macroscopic and microsocopic ingredients was not yet possible. Therefore fission lifetime scaled empirically using Known actinides (Th-Fm).

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Irfu Stability and shell structure (spherical)

- 1949 : The spherical shell model (Mayer, Haxel, Jensen and Suess).
 - 1957 : G. Scharff-Goldhaber "There may be, for instance, another region of relative stability at the doubly magic nucleus $_{126}X^{310}$ "
 - 1966 : Lysekil symposium "Why and how should we investigate nuclei far from the stability line?"



H. Meldner, Ark. Fiz. 36 (1966) 593, shell model → Z=114, N=184

Confirmed by C.Y. Wong PL 21 (1966) 688 (shell model) A. Sobiczewski et al. PL 22 (1966) 500 (Woods-Saxon)

= calculations using phenomenological potentials

Irfu Effective forces

UNIVERSITE PARIS-SACLAY HFB calculations with Skyrme forces : Vautherin 1970
+ Davies 1971, Köhler 1971, Bassichis 1972, Rouben 1972 and 1977, Saunier 1972, Beiner 1974, Brack 1974, Cusson 1976, Vallières 1977, Kolb 1977, Tondeur 1978 and 1980
Spherical calculation for few nuclei, some simplifications
RMF calculations Gambhir 1990, Boersma 1993

 \rightarrow Z= 114 not refuted, although Z = 120, 126 or 138 also suggested

HFB calculations with Gogny force, Berger 1996 : Z=114 not magic !



niversite systematic calculations using self-consistent models (spherical nuclei)

Skyrme forces by Ćwiok, Dobaczewski, Heenen, Magierski and Nazarewicz. NPA 611 (1996) 211

Skyrme and RMF : Rutz, Bender, Bürvenich, Schilling, Reinhard, Maruhn and Greiner, Skyrme and RMF forces. PRC 56 (1997) 238, Bender, Rutz, Reinhard, Maruhn and Greiner PRC 60 (1990) 034304



Spin-orbit splitting



Sirfu Complex nature of SHE

UNIVERSITE PARIS-SACLAY Level density increases Spin orbit \rightarrow orbitals flipped

Low *j* orbitals \rightarrow can modify significantly the gap but not drastically the binding energies \rightarrow smooth island of stability



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Seformed nuclei

UNIVERSITE PARIS-SACLAY First evidence by Schüler and Schmidt (1935) in 151,153 Eu, atomic spectroscopy \rightarrow atomic structure is influenced by the nuclear deformation

Townes systematics 1949 of electric quadrupole moments

1950 : spheroidal model by J. Rainwater, unified model by Bohr and Mottelson

1954 : Nilsson deformed shell model by S.G. Nilsson





rightarrow Irfu Ghiroso systematics of α -decay energies

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pletion in the process of the filling of levels in the simple single-particle shell model may be an oversimplification because this is just the region where the strong surface coupling caused by large spheroidal distortion¹⁰⁻¹³ or configuration interaction^{14–18} of several nucleons may be important. In this connection one might expect on the basis of either the Bohr-Mottelson¹² strong surface coupling model or the de-Shalit-Goldhaber¹⁵ configuration interaction arguments regarding trends of first excited state energies, that if the nucleon configuration at N=152 involves only completely filled levels, the first excited state energies should approach a maximum as is observed in the closing of other shells¹⁹⁻²¹; the experimental evidence so far indicates that this is not the case.^{4,22} Thus it seems that the 152-neutron subshell may be of a fundamentally different nature than the major closed shells.

PR 95 (1954) 293

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Sirfu α-decay energies



SIrfu Harmonic oscillator → Nilsson Model



Irfu The Strutinsky method

Energy = macroscopic + shell correction. NPA 95 (1967) 420



SIrfu Predictions around ²⁵⁰Fm Z=100, N=152

innvorsito



Spectroscopic data vs theory. N=151

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Asai et al. NPA 944 (2015) 308

Irfu Where is the island of stability ?





UNIVERSITE PARIS-SACLAY Fission lifetime calculation : a tremendously difficult task.

1: Which model for shell corrections : phenomenological WS – MHO, effective forces Skryme or RMF ?



2: nuclei explores several degrees of freedom before reaching the saddle point.

3 : fission is a dynamical process; calculation of static energy potentials is not enough.

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https://www.youtube.com/watch?v=DrssJRb301k

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Sendelevium

UNIVERSITE PARIS-SACLAY	New Element Mendelevium, Atomic Number 101*
	A. GHIORSO, B. G. HARVEY, G. R. CHOPPIN, S. G. THOMPSON, AND G. T. SEABORG
	Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California (Received April 18, 1955)
	W E have produced and chemically identified for the first time a few atoms of the element with atomic number 101. Very intense helium ion bombard- ments of tiny targets of 99^{253} have produced a few spontaneously fissionable atoms which elute in the <i>eka</i> -thulium position on a cation resin column.
	253 Es(α ,n) 256 Md target ~ 10 ⁹ atoms, I α ~ 10 ¹⁴ pps, 17 spontaneous fission detected

Last element identified after chemical separation

For heavier elements, breakthroughs needed :

- drop of the cross-section and lifetime
- heavy ion beam needed
- more efficient « physical » separation needed

Sendelevium

UNIVERSITE PARIS-SACLAY	New Element Mendelevium, Atomic Number 101*
	A. GHIORSO, B. G. HARVEY, G. R. CHOPPIN, S. G. THOMPSON, AND G. T. SEABORG
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Irfu The Rf (Z=104) example - Dubna

1964 : G.N. Flerov et al., Dubna

Phys. Lett. 13 (1964) 73



²⁴²Pu(²²Ne,4n)²⁶⁰104 Detection of spontaneous fission using a conveyor belt system Fission detector = glass detector: fission tracks measured offline Spatial distribution of track : implantation-decay correlation and → lifetime Measurement of a 0,3 s fission activity attributed to ²⁶⁰104 (however incorrect interpretation)



Irfu The Rf (Z=104) example - Berkeley

universite Th

The Ghiroso Vertical Wheel. ${}^{249}Cf+{}^{12,13}C \rightarrow {}^{257,259}Rf \rightarrow No$

Parent-daughter correlations : genetic correlations

Detection using Si detectors.

PRL 22 (1969) 1317





Sirfu The VW in detail



Variant using the gas-jet technique (used for the discovery of Sg Z=106) PRL 33 (1974) 1490



Irfu Modern view of genetic correlations





SHIP, GSI. Principle = velocity filter. Typical transmission for Ca+Pb reaction : ~ 30 % Discovery of Z=107-112 by S. Hofmann, G. Münzenberg et al

S. Hofmann



G. Münzenberg

<mark>∕</mark>Irfu ₁₀₇Bh, ₁₀₈Hs, ₁₀₉Mt, ₁₁₀Ds, ₁₁₁Rg, ₁₁₂Cn

UNIVERSITE PARIS-SACLAY 70th : G.S.I.; S.H.I.P. (P. Ambruster); 1975 : first UNIversal Linear ACcelerator beam

- 1981 $_{107}$ Bh (G. Münzenberg *et al.* ZPA 300 (1981) 107) 209 Bi(54 Cr,1n) 262 Bh $\rightarrow ^{258}$ Db $\rightarrow ... \rightarrow ^{250}$ Fm
- 1982 $_{109}$ Mt (G. Münzenberg *et al.* ZPA 309 (1982) 89) 209 Bi(58 Fe,1n) 266 Mt $\rightarrow ^{262}$ Bh $\rightarrow ^{258}$ Db
- 1984 $_{108}$ Hs (G. Münzenberg *et al.* ZPA 318 (1984) 235) 208 Pb(58 Fe,1n) 265 Hs $\rightarrow ^{261}$ Sg $\rightarrow ^{257}$ Rf
- 1994 ₁₁₀Ds, ₁₁₁Rg (S. Hofmann *et al.)* ²⁰⁸Pb(62 Ni,n)²⁶⁹Ds \rightarrow ²⁶⁵Hs \rightarrow ... ZPA 350 (1995) 277 ²⁰⁹Bi(64 Ni,n)²⁷²Rg \rightarrow ²⁶⁸Mt \rightarrow ... ZPA 350 (1995) 281
- 1996 ₁₁₂Cn (S. Hofmann *et al.* ZPA 354 (1996) 229) ²⁰⁸Pb(⁷⁰Zn,1n)²⁷⁷Cn \rightarrow ²⁷³Ds \rightarrow ...

SIrfu **Example of genetic correlations**



needed : total counting rate much larger than Implantation decay rate

Hofmann et al., ZPA 350 (1995) 277

Irfu Position sensitive Si detectors



1980 's : position sensitivity = strips + charge division eg SHIP (picture), RITU DSSD = Double-sided Silicon Strip Detector used in most modern focal plane detectors



Si detector for VAMOS & S3 (GANIL), SHELS (Dubna) 10x10 cm², 128(X)+128(Y) strips

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DGFRS Dubna gas-filled recoil separator (1989) Discovery of elements 114-118 by Oganessian et al.

Typical transmission for Ca+Pb : ~ 45 %





Irfu The principle of a gas-filled separator

UNIVERSITE PARIS-SACLAY lon in a magnetic field : Bρ = Av/q

Charge exchange with the gas : average charge state $<q> = v/v_0 Z^{1/3}$ (Bohr)

 \rightarrow Bp ~ A / Z^{1/3}

 \rightarrow charge state focussing

 \rightarrow no velocity dependence (to first order)

High transmission Target cooling No mass selection Ion slowing down

RITU (Jyväskylä), BGS (Berkeley), DGFRS (Dubna), TASCA(GSI), GARIS (RIKEN), SHANS (Lanzhou), AGFA (ANL), VAMOS-GFS (GANIL soon) He gas used in most cases



Sin the second second



Sirfu DGFRS and Z=118



Sirfu DGFRS and Z=118



SARIS, Riken

Overview of the gas-filled recoil ion separator GARIS **Differential pumping** Rotating ÐÐ target **Detection system** Current monitor **D1** Q1 Q2 D2 Kosuke Morita 元素周期表 元素記号 人工元素 2017 09 28-



Discovery of Nh, Z=113 $^{209}Bi(^{70}Zn,n)^{278}Nh \sigma \sim 22 \text{ fbarn}$ 3 events, 553 days of beam time K. Morita et al. J. Phys. Soc. Jpn. 81 (2012) 103201

Irfu Spectroscopy after alpha decay

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Reminder probability of alpha decay.

Macroscopic part :

 Decay probability increases with Z and Eα, decreases with mass and with transferred angular momentum



- prefers states similar initial and final wave function
- Alpha decay fine structure from 'thorium C' (²¹²Bi discovered in 1929 by S. Rosenblum

C. R. Acad. Sci. 188 (1929) 1401

 Interpretation by G. Gamow (using also gammarays from Black) as population of excited states in the daughter nucleus

Nature 126 (1930) 397

 \rightarrow Alpha decay is a tool for spectroscopy



S. Rosenblum



Gamow, Nature 126 (1930) 397

->∝.

Since the set of the

- **niversite** $0 \rightarrow 0 + \text{transition favoured}$
 - then $0 + \rightarrow 2 + 20 30 \%$



Irfu More complex case (odd nuclei)



Irfu Alpha decay hindrance factor (HF)

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 $\begin{aligned} \mathsf{HF} &= T_{\frac{1}{2}}(\exp.) \ / \ T_{\frac{1}{2}}(\text{theo.}\,, \textit{no nuclear structure, even} - even) \\ T_{\frac{1}{2}}(\exp.) &= \text{partial lifetime of the } \alpha \text{ transition} \end{aligned}$

Empirical HF rules (Loveland, Morrissey and Seaborg : Modern Nuclear Chemistry, Wiley, 2005)

- HF = 1-4 : same initial and final single-particle state
- HF = 4-10 : similar initial and final states
- HF = 10-100 : different single particle states, same parity, same spin projection
- HF = 100-1000 : different single particle states, parity change, same spin projection
- HF > 1000 : different single particle states, parity change, spin flip

✓Irfu ²⁵⁵No as an example

SHIP, GSI. Hessberger et al, EPJA 29 (2006) 165 Choice of the reaction : 208 Pb(48 Ca,1n) 255 No σ ~140 nb, but contaminated by 208 Pb(48 Ca,2n) 254 No σ ~2 µb ²³⁸U(²²Ne,5n)²⁵⁵No σ~100 nb $^{209}\text{Bi}(^{48}\text{Ca},2n)^{255}\text{Lr} \rightarrow (37\%)^{255}\text{No} \sigma \sim 200 \text{ nb}$ Setup = Silicon strip detector $80x35 \text{ mm}^2$, $300\mu\text{m}$ thick + Ge "clover" detector Data a complementary. Cleanest alpha spectra from Ne+U reaction Counts / 10 keV

Samma-rays from ²⁵¹Fm after α decay of ²⁵⁵No











Internal electron conversion

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1911 : Bayer, Hahn and Meitner observe a fine structure in the (β) decay of 'radium B' and 'C' (²¹⁴Pb and ²¹⁴Bi). Phys. Zeit. 12 (1911) 1019

1921 : Ellis. Effect corresponds in 'radium B' to internal electron conversion. Proc. Roy. Soc. Lond. A 99 (1921) 261



- Radiative transition \rightarrow gamma. E(gamma) = E(transition)
- Conversion : electron ejected from the atom E(electron) = E(transition) - E(electron binding energy) Several shells → several electron lines

```
Conversion coefficient \alpha=I(electron)/I(\gamma) \alpha \uparrow when Z \uparrow
\alpha \uparrow when E \downarrow
```
Internal electron conversion



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Example No, E(transition = 200 keV) Bricc code Kibédi et al. NIM A 589 (2008) 202 http://bricc.anu.edu.au/

Shell	E(ce)	E1	M1	E2	M2	E3	M3	E4	M4	E5	M5
Tot		1.188E-01	7.692E+00	1.568E+00	2.757E+01	1.874E+01	1.020E+02	1.535E+02	6.146E+02	1.080E+03	4.387E+03
к	50.70	8.900E-02	5.912E+00	1.294E-01	1.635E+01	1.969E-01	2.305E+01	2.748E-01	2.732E+01	3.661E-01	3.086E+01
L1	170.78	1.216E-02	1.146E+00	9.104E-02	6.275E+00	9.434E-01	2.741E+01	6.779E+00	1.203E+02	4.106E+01	5.371E+02
L2	171.72	6.345E-03	1.773E-01	6.762E-01	9.702E-01	9.326E+00	5.467E+00	7.602E+01	2.972E+01	5.066E+02	1.572E+02
L3	178.15	3.723E-03	3.807E-03	2.642E-01	8.797E-01	2.713E+00	2.175E+01	1.984E+01	2.361E+02	1.247E+02	2.003E+03
L-tot		2.223E-02	1.327E+00	1.031E+00	8.125E+00	1.298E+01	5.463E+01	1.026E+02	3.862E+02	6.724E+02	2.697E+03
M1	192.33	2.856E-03	2.795E-01	2.723E-02	1.683E+00	3.192E-01	8.349E+00	2.541E+00	4.199E+01	1.706E+01	2.153E+02
M2	192.77	1.566E-03	4.871E-02	1.883E-01	2.872E-01	2.801E+00	1.758E+00	2.486E+01	1.044E+01	1.814E+02	6.053E+01
M3	194.27	9.980E-04	1.084E-03	7.944E-02	2.711E-01	8.411E-01	7.190E+00	6.342E+00	8.558E+01	4.139E+01	8.005E+02
M4	194.95	5.586E-05	5.804E-05	1.346E-03	5.283E-03	2.912E-02	1.392E-01	1.566E+00	2.165E+00	3.363E+01	2.572E+01
M5	195.24	5.702E-05	3.116E-05	5.151E-04	2.340E-04	3.581E-02	5.972E-02	1.062E+00	3.531E+00	1.719E+01	7.502E+01
M-tot		5.533E-03	3.294E-01	2.968E-01	2.247E+00	4.026E+00	1.750E+01	3.638E+01	1.437E+02	2.906E+02	1.177E+03

Measurement of conversion coefficient → mulitpolarity. (ambiguous in some cases however) Even better : measurement of conversion on several subshells

Irfu After internal conversion...

Internal conversion

- \rightarrow vacancy in the atomic shell
- \rightarrow rearrangement of the atomic shell followed by electron (Auger, Coster-Kroning) and/or X-ray emission



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Irfu Example (Z=99 conversion 50 keV M1)



1 Conversion L _I	23.2
2 Coster-Kronig L _I -L _{III} M _{III}	1.1
$3 \times L_{III} - M_{V}$	16.0
$4 \times M_{III} - N_{I}$	3.4
5 Auger M _V -N _V N _{VII}	2.9
And so on	

These atomic transitions are emitted in coincidence with the α decay and will (partially) be detected in the implantation detector

 \rightarrow summing



Alpha spectra have to be taken with care !

Simulation (eg Geant4) needed to understand alpha spectra and account properly for the shape of alpha spectra. See eg NIMA 589 (2008) 230





Irfu ²⁵⁵No at JAEA, gas-jet technique









Irfu Conve<mark>rsion ele</mark>ctron detection











Sirfu Offline electron spectroscopy ²⁵⁰Bk



Irfu Identification using X-rays

- 1906 Charles Barkla : X-ray energy is characteristic of an element (\rightarrow nomenclature K, L, M, ...).
- 1913 Henry Moseley. Linear relation between X-ray energy and Z
- \rightarrow rearrange elements according to atomic number
- \rightarrow gaps in gaps in the atomic number sequence at numbers 43, 61, 72, and 75



Charles Barkla

Henry Moseley



∠Irfu X-ray identification of Rf Z=104









Irfu X-ray Identification of Z=115 (?)



Irfu ²⁸⁸115 decay chain at LBNL







Data compatible with <u>NO</u> Bh X-rays.

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- 1917. Isomerism predicted by F. Soddy. Nature 99 (1917) 433
 - 1921. Discovery of isomerism by Otto Hahn.

Decay from 'uranium X2' to 'uranium Z' (²¹⁴Pa isomer decay). Naturwissenschaften 9 (1921) 84

- 1935. Discovery of isomerism in artificial radioactivity (⁸⁰Br) by I. Kurchatov using neutron irradiation
- 1936. Explanation of isomers as spin traps by von Weiszäcker. Naturwissenschaften 24 (1936) 813

"There is no strict half-life requirement for a nuclear excited state to be designated an 'isomer', though it should at least be long lived compared to other states with similar angular momentum and excitation energy" Walker and Xu, Phys. Scr. 91 (2016) 013010

Spin traps, shape isomers, K-isomers





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Decay of a high-K state Selection rule : multipolarity λ of the transition must be larger than Δ K. If not, then transition is forbidden.

In real, transition is not forbidden but hindered. Degree of K forbidness $v = \Delta K - \lambda$

Empirical rule : each degree of forbidness increases the lifetime by a factor of 100 compared to Weisskopf estimates.

Recent review : Walker and Xu, Phys. Scr. 91 (2016) 013010

Κ

Irfu K isomers in heavy nuclei: an old story



MAY 1973

Irfu Transfermium high-K isomer



Irfu Why are high-K isomers interesting ?

universite • In even-even nuclei, 0+ states are trivial. 2qp are not !

- Pair breaking:
$$E_{2qp} = \sqrt{(E_{sp1} - \lambda)^2 + \Delta^2} + \sqrt{(E_{sp2} - \lambda)^2 + \Delta^2}$$

Esp vs fermi level Pairing gap

- \rightarrow pairing correlations
- \rightarrow study of single-particle states





Irfu Why are high-K isomers interesting ?

• Pick experimentally states that would not be accessible otherwise, or with too low intensity

- Spectroscopy of states above the isomer (collectivity)
- States along the decay path
- High-K states may enhance the stability of SHN due to larger fission barrier (anti-fission role).
 Xu et al. PRL 92 (2004) 252501
- Comparison with theory : proper calculation of 2qp state is very complicated.
 - Pairing gap
 - Recoupling
 - Possible role of vibrations and octupole correlations (\rightarrow QRPA)
 - In general agreement is poor in particular for self-consistent models which do not reproduce Z=100 and N=152 deformed shell gaps

Irfu Modern Isomer tagging



<mark> /</mark> Irfu

²⁵⁴No K-isomer 30 years after Ghiroso

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Level scheme and single-particle configuration not (yet) clear

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²⁵⁴ No K-isomers





²⁵⁴ No K-isomers

B



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$$B(E2) = \frac{5}{16\pi} < I K 2 0 | I-2 K >^{2} Q_{0} (e^{2} \text{ fm}^{4})$$

$$(M1) = \frac{3}{4\pi} K^{2} (g_{K} - g_{R})^{2} < I K 1 0 | I-1 K >^{2} (\mu_{n}^{2})$$

$$\mu = \left[g_{R}I + (g_{K} - g_{R}) \frac{K^{2}}{I+1} \right] \mu_{N} \qquad g_{R} \sim Z/A$$

$$g_{K} \sim \frac{1}{K} (g_{S} \Sigma + g_{I} \Lambda)$$

 g_K is characteristic of the orbital(s) and helps to constrain the single-particle alignment.

For 2 qp however no so simple since the g_K factors sum.

Gallagher–Moszkowski rule : coupling anti-parallel spins favoured $\rightarrow g_K \sim \frac{1}{K} g_l(\Lambda_1 + \Lambda_2) = 1$ for protons, 0 for neutrons

Also a good (better) case for prompt spectroscopy



Sirfu Isomers in heavy nuclei

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Also 3qp high-K isomers in even-Z, even-N isotopes

Sirfu Ground state properties

ARIS-SACLAY • Mass measurement

One of the most fundamental quantity in nuclear physics and test for the models

• Laser spectroscopy

Basics = influence of the nucleus on the atomic electrons

Sirfu Mass measurement

• In VHE/SHE : mass usually deduced from alpha decay. Chain anchored to lighter nucleus which mass is known.

- In some SHE decay chain ending by fission (hot fission region)
- Problem in odd nuclei since most intense alpha line not a gs to gs transition.



Sirfu Mass measurement

Recent breakthrough : Mass measurement in ²⁵²⁻²⁵⁵No, ^{255,256}Lr at SHIP + SHIPTRAP (penning trap)



Sirfu Mass measurement in No-Lr






Sirfu Masses vs models





te Mass measurement of isomeric states

Use of an ion trap for purification before spectroscopy

- trap assisted decay spectroscopy
- In-trap decay spectroscopy = detectors in the trap.

→ see eg conversion electron in-trap spectroscopy at REXTRAP (ISOLDE) Weissman et al NIM A 492 (2002) 451, MLLTRAP Weber, P. Müller, P.G. Thirolf Int. J. Mass Spec. 349 (2013) 270

Laser spectrosopy

Basics : effect of the nuclear moments (electric quadrupole, magnetic dipole) and radius on the atomic lines. Nuclear model independent.

- Small effect therefore high precision needed.
- Atom excitation using lasers
- Scan of the laser frequency \rightarrow selective ionisation
- \rightarrow spectroscopy



Spin and parity



Irfu ²⁵³Es optics spectroscopy case (no laser)



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Irfu Laser spectroscopy status (2017/03)



http://www.ikp.tu-darmstadt.de/gruppen_ikp/ag_noertershaeuser/research_wn/exotic_nuclei_wn/uebersicht_2/laserspectroscopy_survey.en.jsp

SIrfu The RADRIS technique at SHIP



Buffer Gas

Filament

(Ta)

PIPS-

Detector



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²³²⁻²⁵⁴No laser spectroscopy

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^{252,254}No:

I = 9/2

M. Laatiaoui et al., Nature 538 (2016) 495 \rightarrow Isotopic shift

- ²⁵³No, M. Laatiaoui et al. to be published
 - Fine structure not fully resolved
 - Compatible with I=9/2
 - µ, Qs

Irfu Hyperfine splitting

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$$\Delta E_{HFS} = \Delta E_{dipole} + \Delta E_{quadrupole}$$
$$\Delta E_{HFS} = \frac{A}{2}C + \frac{B}{4} \frac{\frac{3}{2}C(C+1) - 2IJ(I+1)(J+1)}{IJ(2I-1)(2J-1)}$$

I : nuclear spin; *J*: atomic spin

$$C = F(F+1) - J(J+1) - I(I+1)$$

A, B: hyperfine factor

$$A = \frac{\mu B_e(0)}{IJ} \qquad \mu: \text{ nuclear magnetic dipole moment}$$
$$B = eQ_s \left(\frac{\partial^2 V}{\partial z^2}\right) \qquad Q_s: \text{ nuclear electric quadrupole moment}$$

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Solution Forthcoming facilities, upgrades

UNIVERSITE PARIS-SACLAY • S3 at SPIRAL2/GANIL

• SHE factory, Dubna

GSI cw-linac upgrade





• ATLAS upgrade at ANL

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5

Piot and the S³ collaboration, Acta Phys. Pol. B **43** (2012) 285

SHE factory, Dubna



http://flerovlab.jinr.ru/flnr/she_factory_no.html

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SI LINAC upgrade

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Overall gain \times 40 compared to present facility.

Bath et al. EPJ Web of conferences 138 (2017) 01026

sen -EJC 201

7

Ch. Thei



versite • Synthesis of new nuclei/elements

- Heavier and heavier
- More neutron rich (MNT reactions, etc.)
- Spectroscopy
 - Heavier elements, more details
 - Decay spectroscopy
 - Conversion electrons
 - Trap-assisted, In-trap
 - Prompt spectroscopy
 - Conversion electrons
 - Beyond ²⁵⁶Rf
 - High-K isomers, 2qp, 3qp, 4qp
 - Elements in the U-Es region
- Ground states properties
 - Mass measurements
 - Laser spectroscopy
- Theory

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- the Z=100, N=152 puzzle
- Beyond mean field

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Irfu Naming of the elements

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Naming ceremony conducted at the GSI on 7 September 1992 for the namings of elements 107, 108, and 109 as nielsbohrium, hassium, and meitnerium

Irfu Naming of the elements



Sirfu Naming of the elements





Irfu Naming of the elements

UNIVERSITE PARIS-SACLAY Discovery of elements 104-106 was controversial. Groups who claimed the discovery named these elements.

The situation was clarified in 1997 only by the IUPAC (International Union of Pure and Applied Chemistry).

Procedure :

- Discovery approved by a joint IUPAC–IUPAP Working Group
- Discoverers suggest a name to the IUPAC Inorganic Chemistry Division
- The division examine the proposed name and symbol for suitability
- Public review
- Formal naming

Irfu Naming of the elements

Latest elements approved and named :

- 2003 Z=110 Ds, darmstadtium (GSI)
- 2004 Z=111 Rg, roentgenium (GSI)
- 2010 Z=112 Cn, copernicium (GSI)
- 2012 Z=114 FI, flerovium (Dubna and Livermore)
 Z=116 Lv, livermorium (Dubna and Livermore)
- 2016 Z=113 Nh, nihonium (RIKEN)
 - Z=115 Mc, moscovium (Dubna, Livermore, and Oak Ridge) Z=117 Ts, tennessine (Dubna, Livermore, and Oak Ridge) Z=118 Og, oganesson (Dubna and Livermore)

Elements are universal. Should we name the heaviest elements ?





Further reading

Irfu Holwynium element 120

K.-H. SCHEER L'ÉLÉMENT 120 fleuve noir

Original book in German «Ordnungszahl 120 »

In 2002, element 120 « holwynium » was synthesized by Pr. Holwyn in a US secret atomic base (working on the cobalt bomb) located on the dark side of the moon. This element seems to be useless, but 3400 grams were stolen by Asian enemies. Holwynium has half-life of 2.6 y and is obtained bombarding C on halmanium 112, itself made using a superbevatron. An official from DAS « Département antiespionnage scientifique » is sent to the moon to fix the problem. It turns out that decay of element 120 produces a kind of stable mesons which can be used to produce mesonic atoms of deuterium. Since the radius of these atoms is smaller, controlled fusion is highly favoured. This provides an inexhaustible source of energy. The enemy base on the moon is destroyed using an H bomb.

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université







In French : Tome 8 « La relève » 1ere partie.

Batman DC #45

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General Electrics, 1948

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From Wikipedia : «Robert Scott Lazar (born January 26, 1959) claims to have worked on reverse engineering extraterrestrial technology at a site called S-4, near the Area 51 test facility, and that the UFOs use gravity wave propulsion. This is powered by the, at the time, undiscovered element 115 »







Element 115 + p \rightarrow 116 116 decay \rightarrow 2 antiprotons Antimatter \rightarrow antigravity waves + antigravity amplifiers

http://www.boblazar.com/













RILAC ① 線形加速器 「RILAC」で 原子番号30、 質量数700の 亜鉛(^{*0} 2n)の 原子核を光速 10%にまで加	ど合113 ん成実素 の 速
標的核 ²⁰⁹ [Bi]	 ② その亜鉛ビームを 標的の原子核、 原子番号83、 質量数209の ピスマス(²⁰⁰Bi) に照射する (1秒間に2兆5000億個 の亜鉛の原子核が ピスマスに衝突 する)
励起状態(高温)の 複合核 ²⁷⁹ [113]	③ ビスマスと 亜鉛が完全に 融合して 279113ができる
基底状態の核 278[113]	 ④ ^{2™}113から 中性子1個が 放出され 目的の原子核 である ^{2™}113合成







The Big Bang Theory season 7, episode 6



Further reading (II)



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