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Quest for Superheavy Elements

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Ecole Joliot-Curie "Nucleus through the looking glass – High intensity stable and ISOL beam frontier" La Villa Clythia – Fréjus – France – September 30-October 05, 2012

What's on the menu this week?

-Discovery of the transuranium elements: Z=93 - 112 -Stability of superheavy elements I

Lesson 2:

-Discovery of the transuranium elements: Z=113- ... -Stability of superheavy elements II

-Reactions: synthesis of SHE

-Search for new elements at GSI

What is a superheavy element

Assumptions:

 "... composite nuclear systems that live less than about 10⁻¹⁴ seconds (the generally accepted upper limit for a compound nucleus lifetime) shall not be considered a new element."

B.G. Harvey et al. Science 193 (1976) 1271

2.) "Superheavy Elements" is

 a synonym for
 "elements which only exixts
 because of their (microscopical)
 shell stability."



M. Schädel, 2007

Cross Sections in Hot / Cold / 48Ca Induced Fusion Reactions





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Chart of nuclei September 2012







New Elements from Dubna (⁴⁸Ca+An) ...and news from RIKEN







Cross Sections in Hot / Cold / 48Ca Induced Fusion Reactions



Discovery of the transuranium elements



Superheavy elements 2012: 15 out of 118 (13%!)

Ζ	Name	Symbol
104	Rutherfordium	Rf
105	Dubnium	Db
106	Seaborgium	Sg
107	Bohrium	Bh
108	Hassium	Hs
109	Meitnerium	Mt
110	Darmstadtium	Ds
111	Roentgenium	Rg
112	Copernicium	Cn
113	(yet unnamed)	(uut) / E113
114	Flerovium	FI
115	(yet unnamed)	(uup) / E115
116	Livermorium	Lv
117	(yet unnamed)	(uus) / E117
118	(yet unnamed)	(uuo) / E118

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The periodic table of the elements 2012

Radio elements:

3 R 5 C

8

10 natural radioelements[•] } 38 von 118 (>30%!)

$\frac{18}{18}$										18								
		20	Syr	imei	ic ei	eme	nts	-									2	1
ł	2	(thereof 26 transuranics)									13	14	15	16	17	He	I	
3	4	(there of 4E transcription = 420())									5	6	7	8	9	10	I	
i	Be	(thereof 15 transactinides = 13%)									В	С	N	0	F	Ne	I	
1	12											13	14	15	16	17	18	I
a	Mg	3	4	5	6	7	8	9	10	11	12	AI	Si	P	S	CI	Ar	I
9	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	I
۲	Са	Sc	Ti	V	Cr	Mn	Fe	Со	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr	I
7	38	39	40	41	42	43 •	44	45	46	47	48	49	50	51	52	53	54	I
b	Sr	Y	Zr	Nb	Мо	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те	I	Xe	I
5	56	57+*	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	ł
S	Ва	La	Hf	Та	W	Re	Os	lr	Pt	Au	Hg	TI	-Pb-	Bi	Po	At	Rn	I
7 •	88	89+	104•	105	106•	107•	108•				112•		114•					
r	Ra	Ac	Rf	Db	Sg	Bh	Hs	109	110	111	Cn	113	FI	115	116	117	118	1
_						-		Mt	Ds	Rg					Lv			
		*	58	59	60	61•	62	63	64	65	66	67	68	69	70	71		
			Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Но	Er	Tm	Yb	Lu		
		••	90	91	92	93 •	94•	95 •	96•	97•	98•	99•	100	101	102	105		
			Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr		

Lessons learned

- All heaviest chemical elements have been discovered in physics experiments with recoil separators
- Reports on all elements up to Z=118 published
- Cross sections drop exponentially in "cold (1n)" and "hot (5n)" fusion experiments, but are almost constant from Z=112-118 in ⁴⁸Ca induced reactions

IUPAC rules on how a new element gets named



New Element?

Analysis of the claim by IUPAC/IUPAP

Publication of the analysis in *Pure and* Applied Chemistry

Invitation of the credited group to propose a name

Provisional recommendation presenting the proposed name

Public review

Final approval by the IUPAC Council

Publication of the approved name in *Pure and Applied Chemistry*

Claim from Dubna for element 113

PHYSICAL REVIEW C 69, 021601(R) (2004)

Experiments on the synthesis of element 115 in the reaction ²⁴³Am(⁴⁸Ca, xn)^{291-x}115

Yu. Ts. Oganessian, V. K. Utyonkoy, Yu. V. Lobanov, F. Sh. Abdullin, A. N. Polyakov, I. V. Shirokovsky, Yu. S. Tsyganov, G. G. Gulbekian, S. L. Bogomolov, A. N. Mezentsev, S. Iliev, V. G. Subbotin, A. M. Sukhov, A. A. Voinov, G. V. Buklanov, K. Subotic, V. I. Zagrebaev, and M. G. Itkis *Joint Institute for Nuclear Research*, 141980 Dubna, Russian Federation

J. B. Patin, K. J. Moody, J. F. Wild, M. A. Stoyer, N. J. Stoyer, D. A. Shaughnessy, J. M. Kenneally, and R. W. Lougheed University of California, Lawrence Livermore National Laboratory, Livermore, California 94551, USA (Received 8 September 2003; published 2 February 2004; publisher error corrected 3 February 2004)

Claim from Dubna for element 113





2011 IUPAC/IUPAP assessment of Dubna claim for element 113

113; 03 The collaboration of Oganessian et al. [13]

In 2007, this collaboration investigated the hot fusion of ⁴⁸Ca with ²³⁷Np and reported two four-member α-decay chains commencing at ²⁸²113, passing through ²⁷⁸Rg, ²⁷⁴Mt, and ²⁷⁰Bh, and leading, in just one chain, to ²⁶⁶Db decay by spontaneous fission with a 32 min lifetime. The first two events in each chain showed excellent mutual agreement for both decay energies and lifetimes. The third member gave lifetimes of 470 and 810 ms. None of the nuclides had been previously characterized

JWP ASSESSMENT: The collaborations of Oganessian et al. at Dubna were essentially contemporaneous with those of Morita et al. at RIKEN. The results are encouraging but do not meet the criteria for discovery because of the paucity of events, the lack of connections to known nuclides, and the absence of cross-bombardments.

Claim from RIKEN for element 113 1st paper

Journal of the Physical Society of Japan Vol. 73, No. 10, October, 2004, pp. 2593-2596 ©2004 The Physical Society of Japan

Experiment on the Synthesis of Element 113 in the Reaction ²⁰⁹Bi(⁷⁰Zn,n)²⁷⁸113

Kosuke MORITA^{1*}, Kouji MORIMOTO¹, Daiya KAJI¹, Takahiro AKIYAMA^{1,2}, Sin-ichi GOTO³, Hiromitsu HABA¹, Eiji IDEGUCHI⁴, Rituparna KANUNGO¹, Kenji KATORI¹, Hiroyuki KOURA⁵, Hisaaki KUDO⁶, Tetsuya OHNISHI¹, Akira OZAWA⁷, Toshimi SUDA¹, Keisuke SUEKI⁷, HuShan XU⁸, Takayuki YAMAGUCHI², Akira YONEDA¹, Atsushi YOSHIDA¹ and YuLiang ZHAO⁹

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(Received July 30, 2004)



2011 IUPAC/IUPAP assessment of RIKEN claim for element 113

113; 02 The collaboration of Morita et al. [10,11]

Production of two chains of α -emitting nuclides was reported by Morita et al. from the cold fusion reaction of a bismuth target with a ⁷⁰Zn beam at the RIKEN heavy-ion facility in Japan, the first in 2004 [10] and the second in 2007 [11]. The former study reports the α -chain commencing with ²⁷⁸113 proceeding through ²⁷⁴Rg, ²⁷⁰Mt, ²⁶⁶Bh, and terminating via spontaneous fission decay assigned to ²⁶²Db. All α -energies and lifetimes were measured. In the subsequent study, a very similar sequence was found but with some reproducibility difficulties. The full α -energy for ²⁷⁰Mt was not measured; those for ²⁶⁶Bh were in disagreement (9.08 vs. 9.77 MeV); and the lifetimes for ²⁶²Db spontaneous fission were significantly different (41 vs. 0.8 s). For both chains, position-sensitive detectors were used. These provide a high degree of confidence that the observed decays are indeed sequential decays in each case. Nuclides reported in these chains do not correspond to established systems. But a single report of a triple-coincidence of α -emitters commencing with ²⁶⁶Bh has been described by Wilk et al. [12]. Production was via the hot fusion ²²Ne + ²⁴⁹Bk reaction, and the leading event had an α -particle energy of 9.29 MeV, within the uncertainties of the RIKEN results, with a lifetime of 1–10 s. It was followed by a 28 s α decay, not by spontaneous fission. The latter observation is in contrast to the RIKEN result.

JWP ASSESSMENT: The work of the collaboration of Morita et al. is very promising but has not met the criteria for discovery owing to the paucity of events, the absence of firm connection(s) to known nuclides, and the inconsistencies noted above.

New Result in the Production and Decay of an Isotope, ²⁷⁸113, of the 113th Element

Kosuke Morita^{1*}, Kouji Morimoto¹, Daiya Kaji¹, Hiromitsu Haba¹, Kazutaka Ozeki¹, Yuki Kudou¹, Takayuki Sumita^{2,1}, Yasuo Wakabayashi¹, Akira Yoneda¹, Kengo Tanaka^{2,1}, Sayaka Yamaki^{3,1}, Ryutaro Sakai^{4,1}, Takahiro Akiyama^{3,1}, Shin-ichi Goto⁵, Hiroo Hasebe¹, Minghui Huang¹, Tianheng Huang⁶, Eiji Ideguchi^{7†}, Yoshitaka Kasamatsu^{1‡}, Kenji Katori¹, Yoshiki Kariya⁵, Hidetoshi Kikunaga⁸, Hiroyuki Koura⁹, Hisaaki Kudo⁵, Akihiro Mashiko¹⁰, Keita Mayama¹⁰, Shin-ichi Mitsuoka⁹, Toru Moriya¹⁰, Masashi Murakami⁵, Hirohumi Murayama⁵, Saori Namai¹⁰, Akira Ozawa¹¹, Nozomi Sato⁹, Keisuke Sueki¹¹, Mirei Takeyama¹⁰, Fuyuki Tokanai¹⁰, Takayuki Yamaguchi³, and Atsushi Yoshida¹

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		Table				
New Result in 1 Kosuke Morita ^{1*} Takayuki Sumr	year	Beamtime month/day	Irradiation time (days)	Beam dose/sum (×10 ¹⁹)	Number of observed events	e 113th Element кі ¹ , Yuki Kudou ¹ ,
Rvutaro Sak	2003	9/5-12/29	57.9	1.24/1.24	0	ghui Huang ¹ .
Tianheng Hu	2004	7/8-8/2	21.9	0.51/1.75	1	shiki Kariya ⁵ ,
Hidetoshi Kıkı	2005	1/20-1/23	3.0	0.07/1.82	0	ita Mayama ¹⁰ ,
Shin-ichi MITSU	2005	3/20-4/22	27.1	0.71/2.53	1	⁵ , Saori Nама1 ¹⁰ ,
Akira Oza	2005	5/19-5/21	2.0	0.05/2.58	0	Tokanai ¹⁰ ,
	2005	8/7-8/25	16.1	0.45/3.03	0	
¹ DIKEN	2005	9/7-10/20	39.0	1.17/4.20	0	08 Japan
$^{2}Faculty$	2005	11/25-12/15	19.5	0.63/4.83	0	10, Japan
	2006	3/14-5/15	54.2	1.37/6.20	0	
	2008	1/9-3/31	70.9	2.28/8.48	0	
⁶ Inst	2010	9/7-10/18	30.9	0.52/9.00	0	China
	2011	1/22-5/22	89.8	2.01/11.01	0	m
⁸ Rese	2011	12/2-12/19	14.4	0.33/11.34	0	Japan
	2012	1/15-2/9	25.0	0.56/11.90	0	
	2012	3/13-4/17	33.7	0.79/12.69	0	
(Receiv	2012	6/12-7/2	15.7	0.25/12.94	0	7, 2012)
(HUUU)	2012	7/14-8/18	32.0	0.57/13.51	1	,, 2012)
	Total		553	13.51	3	

	Chain 1 ¹⁾	Chain 2 ²⁾	Chain 3 (present)		of beamti	me used.					
	E (MeV)	E (MeV)	E (MeV)	Assignmen	Rh 260	Rh 261	Rh 262		Bh 264	Bh 264	Rh 266
	ΔT	ΔT	ΔT	mean lifeti	25 mg	11.º ma			0.07 c	BIT 20.	110
	Position	Position	Position		α 10.16	α~10.0; e	α 10.37; α 10.08; 10.20 9.69		α.9.48; 9.62;	α 9.24	α 9.08; 9.29
	36.75	36.47	41.91	3	Sg 259	Sg 260	Sa 261	Sa 262	Sa 263	N o	Sa 265
ER	44.6 ns (TOF)	45.7 ns (TOF)	42.6 ns (TOF)		0.28 s	4.95 ms	184 ms	15 ms		r /	<> 8.5.5 14.4.5
	30.3 mm	30.1 mm	4.9 mm		α 9.59, 9.01- 9.47; sf≦12%; γ	α 9.75; 9.72; sf (71±3%)	α 9.56; 9.52; 9.47	sf; α≦16%	α 9.05 α 9.25; st st<5% (13±6%)	13	a 8.84 a 8.69
	11.68 (0.04)	11.52 (0.04)	11.82 (0.06)	²⁷⁸ 113	Db 258	Db 259	Db 260	Db 261	Db 262	Db 263	
α_1	0.344ms	4.93 ms	0.667 ms	2.0 ^{+2.7} _{-0.7} ms		0.51 s	1.5 s	1.8 s	34 s	27 s	
	30.5 mm	30.2 mm	4.4 mm	a ; :	α 9.17 e α 9.20;e? γ221	α9.47	α 9.04; 9.12; e/sf?	a 8.93; sf (??)	α 8.45; 8.53; ,8.67; ∈/sf (33%)	9.30; sf (57%); (2+4_%)	
	11.15 (0.07)	11.31 (0.07)	10.65 (0.06)	²⁷⁴ Rg	Rf 257	Rf 258	Rf 259	Rf	Rf 261	Rf 262	Rf 263
α_2	9.26 ms	34.3 ms	9.97 ms	18^{+24}_{-7} ms	180µc 4.1 c 7.2 c	14 ms	2.5 s	ľ /	€8 s 2.6	47ms? 2.1s?	~8 s
	30.4 mm	29.6 mm	4.8 mm	a.75	α8.2-8.5 e(11%) α.9.02; s12%) hy 8.97 y 117.	sf α 9.05 (31±11%)	α 8.77; 8.87; e(15±4%)		α 8.30; α 8.51; sf<11% sf(82%)	sf? sf;α≦3%	
	10.03 (0.07)	2.32 (escape)	10.26 (0.07)	²⁷⁰ Mt	Lr 256	Lr 257	Lr 258	Lr 259	Lr 260	Lr 261	Lr 262
α_3	7.16 ms	1.63 s	444 ms	$0.69^{+0.95}_{-0.26}$ s	25.9 s	0.65 s	3.9 s	6.14 s	3 m	39 m	3.6 h
	29.8 mm	29.5 mm	5.1 mm	(73; 12; 269	α 8.43; 8.52; 8.39; γ 163	α 8.86; 8.80	a 8.595; 8.621; 8.565; 8.654	8.445 (25%)	α 8.03	sf ∈?	-
	9.08 (0.04)	9.77 (0.04)	9.39 (0.06)	²⁶⁶ Bh	No 255	No	No 257	No 258	No 259	No 260 ?	
α_4	2.47 s	1.31 s	5.26 s	3.0 ^{+4.2} _{-1.1} s	3.1 m	t 💉	24.5 s	1.2 ms	59 m	106 ms	
	30.9 mm	29.7 mm	4.9 mm	(0%) 1.2% -	y 187;e≒∈(39%)		α 8.22; 8.32; sf≦1.5%	sf; α≦??	α.7.520; 7.551; (25%); sf≦10%	sf;α≦??	
	204 (SF)	192 (SF)	8.63 (0.06)	²⁶² Db	Md 254	Md 255	Md 256	Md 257	Md 258	Md 259	Md 260
F/α_5	40.9 s	0.787 s	126 s	56 ⁺⁷⁷ ₋₂₁ s	10 m 28 m	27 m	1.30 h	5.52 h	57 m 51.5 d 46.718, 8.763	95 m	31.8 d
	30.3 mm	30.5 mm	4.5 mm		• •	2(82%);SI21.4%	e(91%); st52.8% α7.221;7.155; γ	e(85%); sr≥1%; α7.074; γ 371	arid0%; y369,448; α<1.2%;β* β; <30% (af,e,β):0.3%	sf	sf
			8.66 (0.06)	²⁵⁸ Lr	Fm 25	Fm 254	Fm 255	Fm 256	Fm 257	Fm 258	Fm 259
α_6	_	_	3.78 s	$3.8^{+18}_{-1.7}$ s	3.0 d	3.24 h	20.1 h	70 ns 2.63 h	100.5 d	0.37 ms	1.5 s
			4.7 mm		6.673 γ 272.	sf; γ (99;43);e	sf; γ (81;58);er	k 862; 231 sf (91.9%) sf (??%) a 8.917	γ 242; 180; e	sf	sf

The element 113/115 decay chains from Dubna



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TAsca Small Image mode SPECtroscopy



Dirk Rudolph (Lund University) X-ray Fingerprinting of E115 Decay Chains

TAsca Small Image mode SPECtroscopy





Dirk Rudolph (Lund University) X-ray Fingerprinting of E115 Decay Chains



 Detailed spectroscopy towards Z=108/N=162 (²⁷⁰Hs) → Input for SHE Island of Stability: **114** vs. **120** vs. **126** Fingerprinting SHE: Z identification via characteristic X-rays

200

Lessons learned

- Elements up to 112, and 114/116 named.
- All elements up to 118 claimed. 113/115/117/118 not approved
- No atomic number directly measured for any element beyond 112 (113...)
- New (last week) data on E113 from RIKEN

Stability of SHE

The atomic mass / binding energy Alpha decay

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Mass ↔ Energy

A nucleus with Z protons and N neutrons is lighter than Z protons + N neutrons

 \rightarrow Mass is converted to (binding) energy

The mass of a nucleus (or, an atom) gives direct information on its stability!

Relation between mass and energy:

 $E[J] = m[kg] \cdot (c[m/s])^2$

Atomic masses are given in the unit "u" (unified atomic mass)

Definition: $M(^{12}C) \equiv 12 u$

Using c=299'792'458 m/s gives:1 MeV/c² $\stackrel{\wedge}{=}$ 1.073533·10⁻³ u and 1 u $\stackrel{\wedge}{=}$ 931.494 MeV/c².

(Calculations "in energy": [MeV] or [J]; Calculations "in mass": [MeV/c²] or [kg])



Atomic masses

$M(Z,A) = Z \cdot M_{H} + (A-Z) \cdot M_{N} - BE/c^{2}$

M(Z,A): Mass of the atom (MeV/c²)

- Z: Protonen number
- A: Mass number
- M_{H} : Mass of neutral H atom (938.791 MeV/c²=1.007823 u)
- M_{N} : Masse of neutron (939.573 MeV/c²=1.008662 u)
- BE: Binding energy (MeV)

The semi-empirical mass formula (SEMF)



The semi-em	pirical mass f	formu	la (SEMF)		
BE consists of five cont	Example: BE of ¹² C				
$E_v = a_v \cdot A$	a _v = 15.85 MeV/c²	E _v : E _s : E _c :	190.20 MeV/c ² -96.13 MeV/c ² -11.16 MeV/c ²		
$E_{s} = a_{s} \cdot A^{2/3}$	$a_o = -18.34 \text{ MeV/c}^2$	E _a : E _p :	0.00 MeV/c ² 3.31 MeV/c ²		
$E_{c} = a_{c} \cdot Z^{2}/A^{1/3}$	$a_{c} = -0.71 \text{ MeV/c}^{2}$	BE: M(¹² C):	86.22 MeV/c ² 12.00636 u		
$E_a = a_a \cdot (Z - A/2)^2 / A$	a _a = -92.86 MeV/c ²	\rightarrow why not	t12.0000 u ?		
		Example: BE of ²⁶¹ Rf			
$\int +\delta$ for e,e-nuclei		E _v :	4136.85 MeV/c ²		
$E_p = \{ 0 \text{ for odd-A nucle} \}$	ei	E _s :	-749.02 MeV/c ²		
$-\delta$ for 0,0-nuclei		E _c :	-1201.65 MeV/c ²		
		E _a :	-249.85 MeV/c ²		
$\delta = a_p \cdot A^{-1/2}$	a _p = 11.46 MeV/c ²	E <u>p</u> :	0.00 MeV/c ²		
	r	BE:	1936.33 MeV/c ²		
Average deviation	M(²⁶¹ Rf):	261.095 u			
experimental masse	for compariso	n: AME2003: 261.109 u			

The α decay

Example: ${}^{248}Cm \rightarrow {}^{244}Pu + {}^{4}He$

 $\Delta(^{248}\text{Cm}) = \Delta(^{244}\text{Pu}) + \Delta(^{4}\text{He}) + Q$

 $(\Delta(Z,A) [MeV/c^2] = M(Z,A)[u] - A[u]$ is the mass excess)

Decay energy: Q_{α} [MeV]= { Δ (²⁴⁸Cm) - [Δ (²⁴⁴Pu) + Δ (⁴He)]}c²

 Q_{α} is distributed between the α particle and the daughter. From momentum conservation follows the energie of the α particle:

$$E_{\alpha}\left(^{248}Cm\right) = Q_{\alpha} \cdot \frac{M\left(^{244}Pu\right)}{M\left(^{248}Cm\right)}$$

and the recoil energy of the daughter

$$E_{D}(^{248}Cm) = Q_{\alpha} \cdot \frac{M(^{4}He)}{M(^{248}Cm)}$$

Cs-Isotopes: mass in different mass models



Prediction of E_{\alpha}

Example: ${}^{248}Cm \rightarrow {}^{244}Pu + {}^{4}He$

 $\mathsf{Q}=\{ \Delta(^{248}\mathsf{Cm})-[\Delta(^{244}\mathsf{Pu})+\Delta(^{4}\mathsf{He})] \} \mathsf{c}^2$

	SEMF	AME 2003
∆(²⁴⁸ Cm):	53.485 MeV/c ²	67.392±0.005 MeV/c ²
∆(²⁴⁴ Pu):	45.658 MeV/c ²	59.806±0.005 MeV/c ²
∆(⁴ He):		2.42491565±0.0000006 MeV/c ²
Q_{α} (²⁴⁸ Cm)	5.402 MeV	5.16173 ±0.00025 MeV
E_{α} (²⁴⁸ Cm)	5.315 MeV	5.08 MeV
	•	

 $^{248}\text{Cm}\ \alpha\text{-decay}$ energy of the simple SEMF is precise to about 5%

Nuclear Physics 101: α **-Decay**



Geiger-Nuttall's rule in the region of the heaviest elements



Oganession, Radiochim. Acta 2012

Realististic estimate of stability of superheavy elements – including the border of the Periodic Table – requires good knowledge of nuclear masses

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Experimental weighing of the atom



$E=mc^2$

The perfect balance tells us about nuclear binding energy \rightarrow stability of a nucleus

Shell structure from masses



$$S_{2n}(Z,N) = m(Z,N-2) - m(Z,N) + 2m_n$$

M. Block

Principle of a Penning trap



Penning traps for most highly precise atomic mass measurements





M. Block

LETTERS

Direct mass measurements above uranium bridge the gap to the island of stability

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Direct Mapping of Nuclear Shell Effects in the Heaviest Elements

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