Fission and the discovery of isotopes

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Isotope Day - 4 December 2013

Isotopes were introduced to the world in a letter to the journal 'Nature', published on 4 December 1913 by University of Glasgow chemist Frederick Soddy.

He realised that a single chemical element could occur as atoms with different atomic weights, with different nuclear properties, such as radioactive half-life. He thus reconciled the periodic table with the newly-discovered phenomena of radioactivity, and atomic transformation. He later received the Nobel Prize in Chemistry for this work.

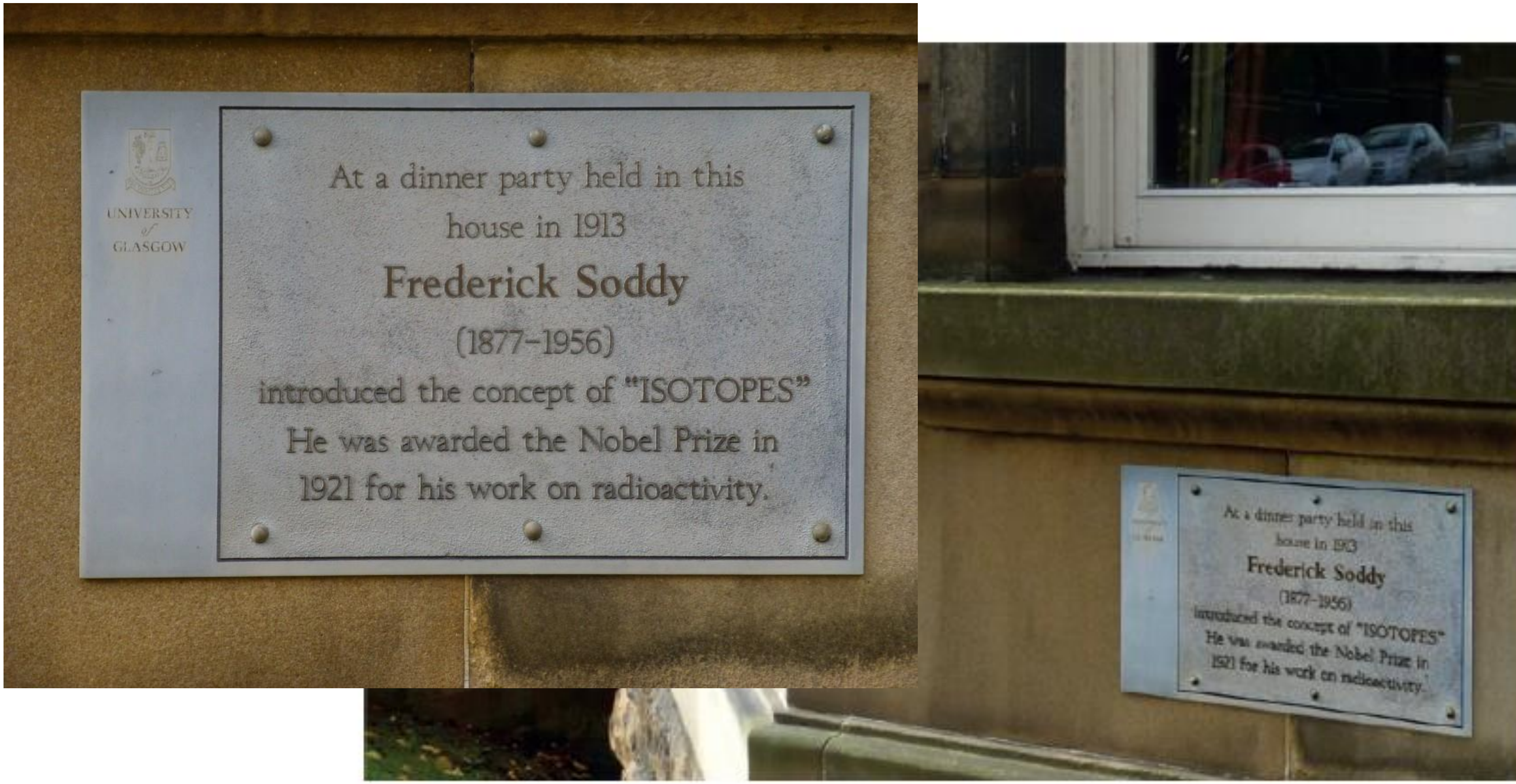
The word 'isotope' itself had been suggested to him by Margaret Todd, a Glasgow GP, during a dinner at 11 University Gardens. Isotope science was truly born at the University of Glasgow.

100-years of isotopes

http://www.gla.ac.uk/hunterian/visit/events/headline_296351_en.html

Origin of the term isotope

http://blogs.nature.com/thescepticalchymist/2013/11/isotope-day.html



B. F. Thornton and Shawn C. Burdette, Nature Chemistry **5** (2013) 979

December 4: Isotope day

DECEMBER 4, 1913] NATURE 399

LETTERS TO THE EDITOR.

growing ova by nurse cells, the latter being phagocytes which capture other cells and stuff them into the

So far as I personally am concerned, this has resulted in a great clarification of my ideas, and it may be helpful to others, though no doubt there is little originality in it. The same algebraic sum of the positive and negative charges in the nucleus, when the arithmetical sum is different, gives what I call "isotopes" or "isotopic elements," because they occupy the same place in the periodic table. They are chemically identical, and save only as regards the relatively few physical properties which depend upon atomic mass directly, physically identical also. Unit changes of this nuclear charge, so reckoned algebraic-

the action of the enzyme which caused it, but also that if these products reached a certain concentration, the enzyme instead of producing further hydrolysis

(NATURE, November 27, p. 372), is strongly supported by the recent generalisation as to the radio-elements and the periodic law. The successive expulsion of one

Exploration of rare isotopes

"Owing to the rapid advance in research on disintegration and the theory of nuclear structure, the existence or non-existence of rare isotopes has acquired an entirely unexpected importance and calls for a short review of their present situation."

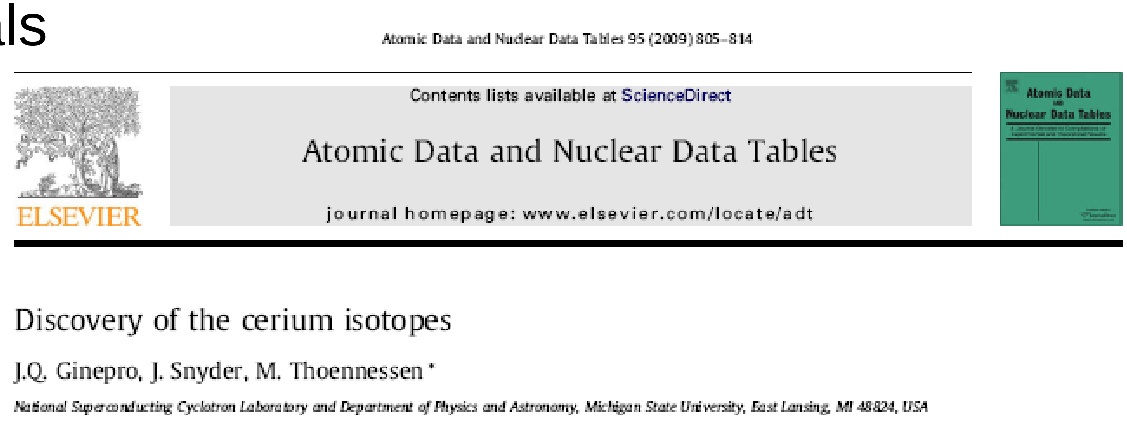
F.W. Aston, Nature **137**, 613 (1936)

Discovery project

- Important to document the history
- Contrary to common perception not easily accessible
- Comprehensive compilation is only now possible

Criteria:

- Clear identification, either through decay-curves and relationships to other known isotopes, particle or γ-ray spectra, or unique mass and Z-identification
- Publish in refereed journals
- Not as strict as dis-covery of elements
- First observation until proven incorrect



Atomic Data and Nuclear Data Tables 95 (2009) 805–814

Contents lists available at ScienceDirect

Atomic Data and Nuclear Data Tables

journal homepage: www.elsevier.com/locate/adt

ELSEVIER

Discovery of the cerium isotopes

J.Q. Ginepro, J. Snyder, M. Thoennessen *

National Superconducting Cyclotron Laboratory and Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824, USA

Discovery of transuranium elements?

Possible Production of Elements of Atomic Number Higher than 92

By PROF. E. FERMI, Royal University of Rome

Nature, June 16, 1934

E. Fermi, Nobel Lecture, December 12, 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. It is known that O. Hahn and L. Meitner have investigated very carefully and extensively the decay products of irradiated uranium, and were able to trace among them elements up to the atomic number 96.*

* The discovery by Hahn and Strassmann of barium among the disintegration products of bombarded uranium, as a consequence of a process in which uranium splits into two approximately equal parts, makes it necessary to reexamine all the problems of the transuranic elements, as many of them might be found to be products of a splitting of uranium.

Early skeptics:

Uber das Element 93.

Von Dr.-Ing. IDA NODDACK, Berlin.

Angew. Chemie 47 (1934) 653

Der Beweis, daß das neue Radioelement die Ordnungszahl 93 hat, ist also noch keineswegs geglückt, da *Fermi* ihn nur durch ein unvollkommen durchgeführtes Ausschlußverfahren versucht hat.

Man kann ebensogut annehmen, daß bei dieser neuartigen Kernzertrümmerung durch Neutronen erheblich andere „Kernreaktionen“ stattfinden, als man sie bisher bei der Einwirkung von Protonen- und α-Strahlen auf Atomkerne beobachtet hat. Bei den letztgenannten Bestrahlungen findet man nur Kernumwandlungen unter Abgabe von Elektronen, Protonen und Heliumkernen, wodurch sich bei schweren Elementen die Masse der bestrahlten Atomkerne nur wenig ändert, da nahe benachbarte Elemente entstehen. Es wäre denkbar, daß bei der Beschießung schwerer Kerne mit Neutronen diese Kerne in mehrere g r ö ß e r e Bruchstücke zerfallen, die zwar Isotope bekannter Elemente, aber nicht Nachbarn der bestrahlten Elemente sind.

The proof that the new radioelement has Z = 93, has not been established…

It is conceivable that… these nuclei decay into several larger pieces…

Apparent confirmation, but…

Über die Umwandlungsreihen des Urans, die durch Neutronenbestrahlung erzeugt werden.

Von **L. Meitner, O. Hahn** und **F. Strassmann.**

Mit 3 Abbildungen. (Eingegangen am 14. Mai 1937.)

Z. Phys. 106 (1937) 249

$$1.\quad U + n \longrightarrow {}^{10''}_{92}U \xrightarrow{\beta} {}^{2,2'}_{93}\text{Eka Re} \xrightarrow{\beta} {}^{59'}_{94}\text{Eka Os} \xrightarrow{\beta} {}^{66\,h}_{95}\text{Eka Ir} \xrightarrow{\beta} {}^{2,5\,h}_{96}\text{Eka Pt} \xrightarrow{\beta} {}_{97}\text{Eka Au ?}$$

$$2.\quad U + n \longrightarrow {}^{40''}_{92}U \xrightarrow{\beta} {}^{16'}_{93}\text{Eka Re} \xrightarrow{\beta} {}^{5,7\,h}_{94}\text{Eka Os} \xrightarrow{\beta} {}_{95}\text{Eka Ir ?}$$

$$3.\quad U + n \longrightarrow {}^{23'}_{92}U \xrightarrow{\beta} {}_{93}\text{Eka Re ?} \longleftarrow {}^{239}U$$

Also müssen die Prozesse Einfangprozesse des Uran 238 sein, was zu drei isomeren Kernen Uran 239 führt. Dieses Ergebnis ist mit den bisherigen Kernvorstellungen sehr schwer in Übereinstimmung zu bringen.

This result is hard to understand within the current understanding of nuclei.

Discovery of ^{140}Ba

Atomic Data and Nuclear Data Tables 96 (2010) 749–758

Discovery of the barium isotopes

A. Shore, A. Fritsch, J.Q. Ginepro, M. Heim, A. Schuh, M. Thoennessen *

2.24. ^{140}Ba

In the 1939 paper, "Nachweis der Entstehung aktiver Bariumisotope aus Uran und Thorium durch Neutronenbestrahlung; Nachweis weiterer aktiver Bruchstücke bei der Uranspaltung" Hahn and Strassmann identified ^{140}Ba for the first time at Berlin–Dahlem in Germany [43]. ^{140}Ba was produced by irradiating Uranium with neutrons from a Ra–Be-neutron source. Decay curves were measured following chemical separation. A previously reported 300 h activity originally labeled as "Ra IV" [44,45], now identified as the fission product "Ba IV," was again observed. Based on the measured half-life of the daughter activity, it was tentatively assigned to ^{140}Ba: "Was die anderen Barium isotope...

O. Hahn, F. Strassmann, Naturwiss. 27 (1939) 89

OH,FS, Nw 26 (1938) 755
OH,FS, Nw 27 (1939) 11

December 22,1938:

Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans mittels Neutronen entstehenden Erdalkalimetalle[1].

Von O. HAHN und F. STRASSMANN, Berlin-Dahlem.

Naturwiss. 27 (1939) 11

Was die „Trans-Urane“ anbelangt, so sind diese Elemente ihren niedrigeren Homologen Rhenium, Osmium, Iridium, Platin zwar chemisch verwandt, mit ihnen aber nicht gleich. Ob sie etwa mit den noch niedrigeren Homologen Masurium, Ruthenium, Rhodium, Palladium chemisch gleich sind, wurde noch nicht geprüft. Daran konnte man früher ja nicht denken. Die Summe der Massenzahlen Ba + Ma, also z. B. 138 + 101, ergibt 239!

If they correspond to technetium, ruthenium, rhodium, palladium has not been tested. One could not have thought about this earlier. The sum of the Ba+Ma mass numbers (128+101) is 239!

Als Chemiker müßten wir aus den kurz dargelegten Versuchen das oben gebrachte Schema eigentlich umbenennen und statt Ra, Ac, Th die Symbole Ba, La, Ce einsetzen. Als der Physik in gewisser Weise nahestehende „Kernchemiker“ können wir uns zu diesem, allen bisherigen Erfahrungen der Kernphysik widersprechenden, Sprung noch nicht entschließen. Es könnten doch noch vielleicht eine Reihe seltsamer Zufälle unsere Ergebnisse vorgetäuscht haben.

As chemist we should rename Ra, Ac, Th to Ba, La, Ce. As “nuclear chemists” close to physics, we cannot take this step, because it contradicts all present knowledge of nuclear physics.

January 28, 1939: Discovery of ^{140}Ba

Nachweis der Entstehung aktiver Bariumisotope aus Uran und Thorium durch Neutronenbestrahlung; Nachweis weiterer aktiver Bruchstücke bei der Uranspaltung[1].

Von OTTO HAHN und FRITZ STRASSMANN, Berlin-Dahlem.

A. Endgültiger Beweis für das Entstehen von Barium aus dem Uran.

In einer vor kurzem in dieser Zeitschrift erschienenen Mitteilung[1] haben wir angegeben, daß die bei der Bestrahlung des Urans mittels Neutronen entstehenden, anfangs für Radiumisotope gehaltenen

[1] Aus dem Kaiser Wilhelm-Institut für Chemie in Berlin-Dahlem. Eingegangen am 28. Januar 1939.

[1] O. HAHN u. F. STRASSMANN, Naturwiss. 27, 11 (1939).

Conclusive proof for the formation of barium from uranium

FEB. 11, 1939 NATURE

Disintegration of Uranium by Neutrons: a New Type of Nuclear Reaction

On the basis, however, of present ideas about the behaviour of heavy nuclei[6], an entirely different and essentially classical picture of these new disintegration processes suggests itself. On account of their close packing and strong energy exchange, the particles in a heavy nucleus would be expected to move in a collective way which has some resemblance to the movement of a liquid drop. If the movement is made sufficiently violent by adding energy, such a drop may divide itself into two smaller drops.

Jan. 16.

LISE MEITNER.
O. R. FRISCH.

Meitner and Frisch were first with the explanation!

May 11, 1939: Discovery of ^{127}Sb and ^{129}Sb

An Investigation of the Products of the Disintegration of Uranium by Neutrons

PHILIP ABELSON
Radiation Laboratory, University of California, Berkeley California
(Received May 11, 1939)

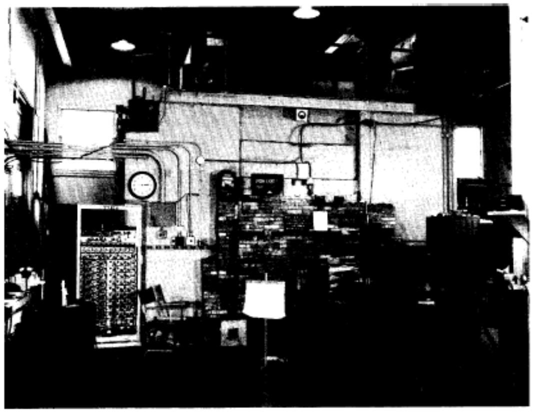
When news of the discovery of the uranium cleavage reached this laboratory.

METHOD

Uranium samples were activated by neutrons formed through the bombardment of beryllium by 8.0-Mev deuterons produced by the cyclotron.

P. Abelson, Phys. Rev. 55 (1939) 876

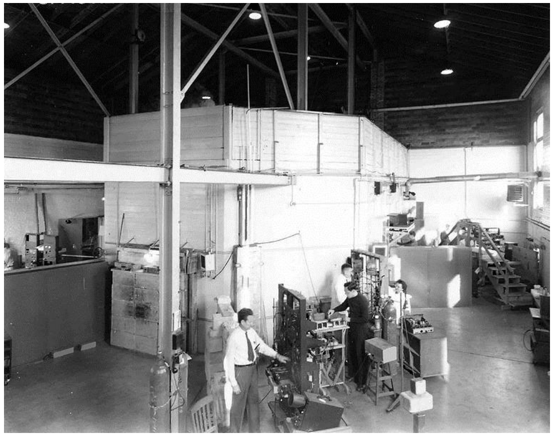
Neutron fission in reactors



Los Alamos homogeneous pile

Clinton Pile (ORNL)

Chicago Pile-3



National Nuclear Energy Series

World War II

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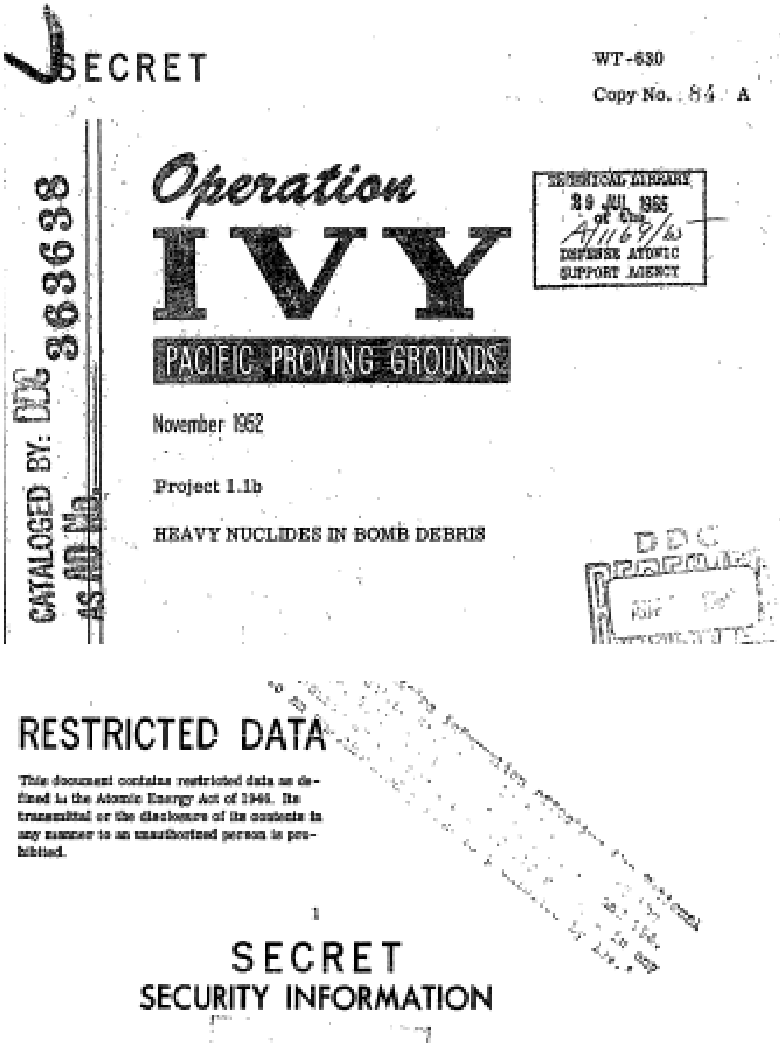

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HEAVY NUCLIDES IN BOMB DEBRIS

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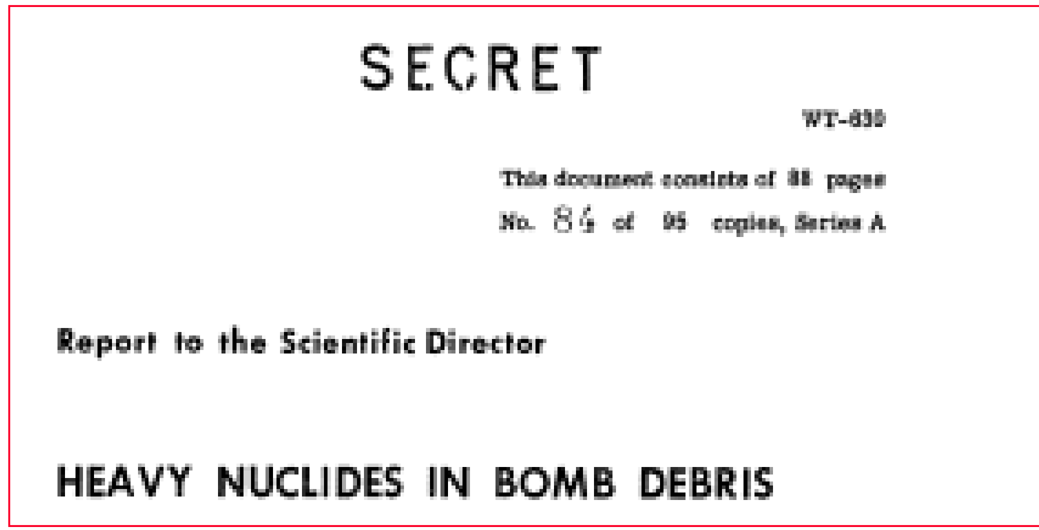

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Report to the Scientific Director

HEAVY NUCLIDES IN BOMB DEBRIS

Classified research

<u>Neptunium and plutonium:</u>

These first two transuranium elements were referred to simply as "element 93" and "element 94" or by code names, …

Throughout 1941, element 94 was referred to by the code name of "copper", which was satisfactory until it was necessary to introduce the element copper into some of the experiments. This posed the problem of distinguishing between the two.

For a while, the plutonium was referred to as "copper" and the real copper as "honest-to-God copper."

"The elements beyond uranium",
G.T. Seaborg and W.D. Loveland (Wiley1990)

Different times

At the suggestion of Dr. J. G. Hamilton and with his aid we have injected known amounts of the supposed eka-iodine into two hyperthyroid guinea pigs, on the chance that it might behave like iodine and be concentrated in the thyroid. The guinea pigs were killed about 4.5 hours after administration of the radioactive material and various portions of the bodies were examined for activity. In one animal the thyroid contained roughly 100 times as much activity as equal masses of other portions of the body.

D.R. Corson *et al.*, Phys. Rev. **57** (1940) 459

…more relaxed!!!

McMillan[10] found a long-lived soft radiation from metal scraped from inside the cyclotron vacuum chamber and suggested it might be due to C^{14} formed by the reaction

$$D_1^2 + C_6^{13} \rightarrow C_6^{14} + H_1^1 + Q_2. \quad (2)$$

Unfortunately, the sample was accidentally lost before any chemistry was performed.

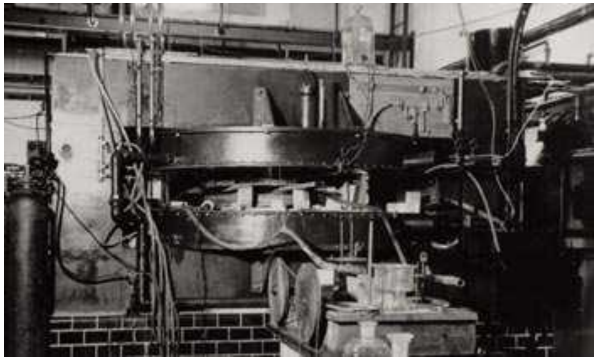
S. Ruben *et al.*, Phys. Rev. **59** (1941) 349

1951: Discovery of ^{90}Kr, ^{90}Rb and ^{91}Rb

Short-Lived Krypton Isotopes and Their Daughter Substances

O. KOFOED-HANSEN AND K. O. NIELSEN
Institute for Theoretical Physics, University of Copenhagen, Copenhagen, Denmark
(Received February 9, 1951)

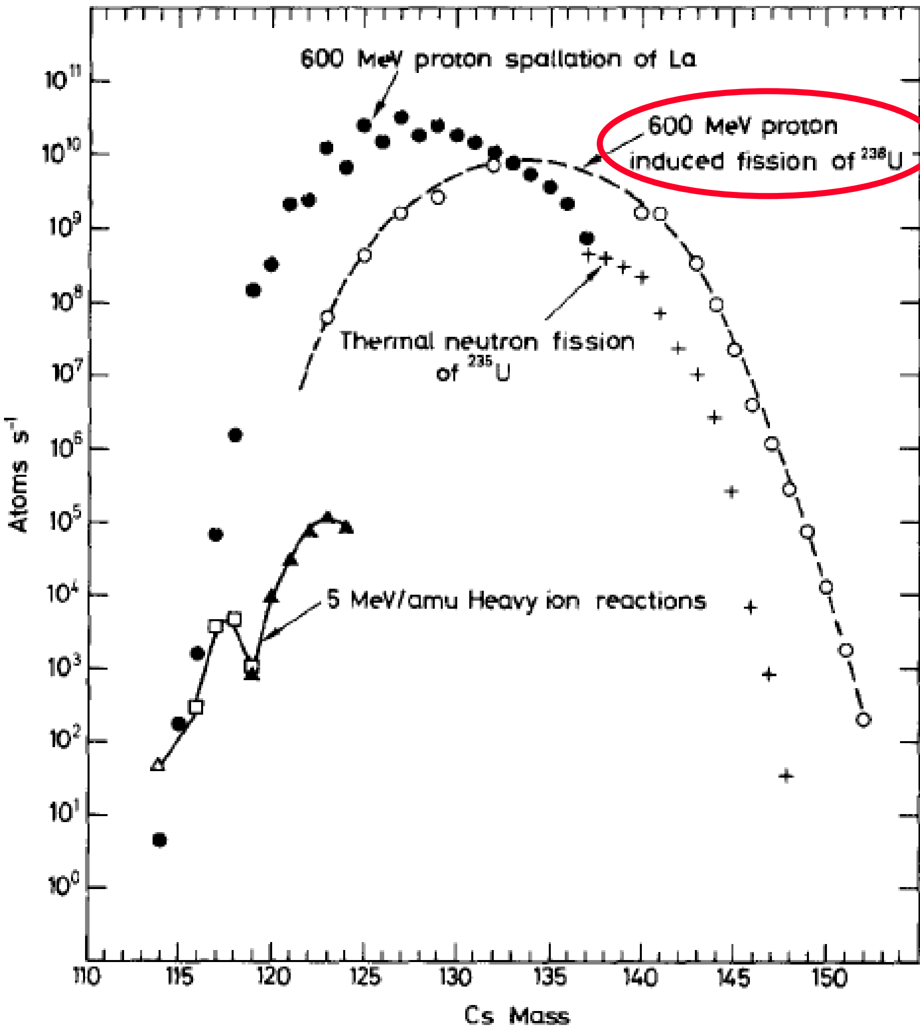
THE isotopes Kr^{89}, Kr^{90}, Kr^{91}, and their daughter substances have been investigated. Krypton formed in fission of uranium was pumped through a 10-m long tube directly from the cyclotron into the ion source of the isotope separator. The



First application of ISOL

O. Kofoed-Hansen and K.O. Nielsen, Phys. Rev. 82 (1951) 86

1979: Discovery of $^{148-152}$Cs

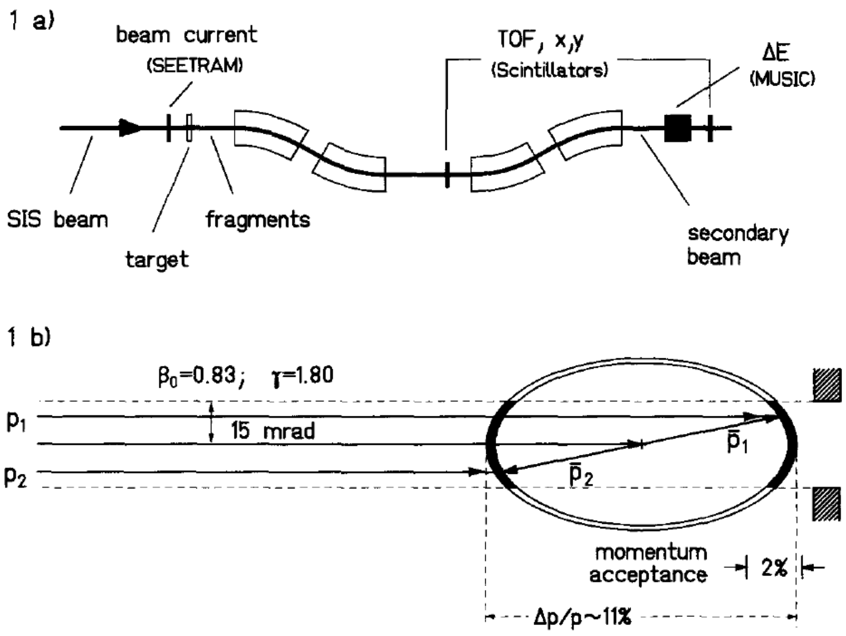


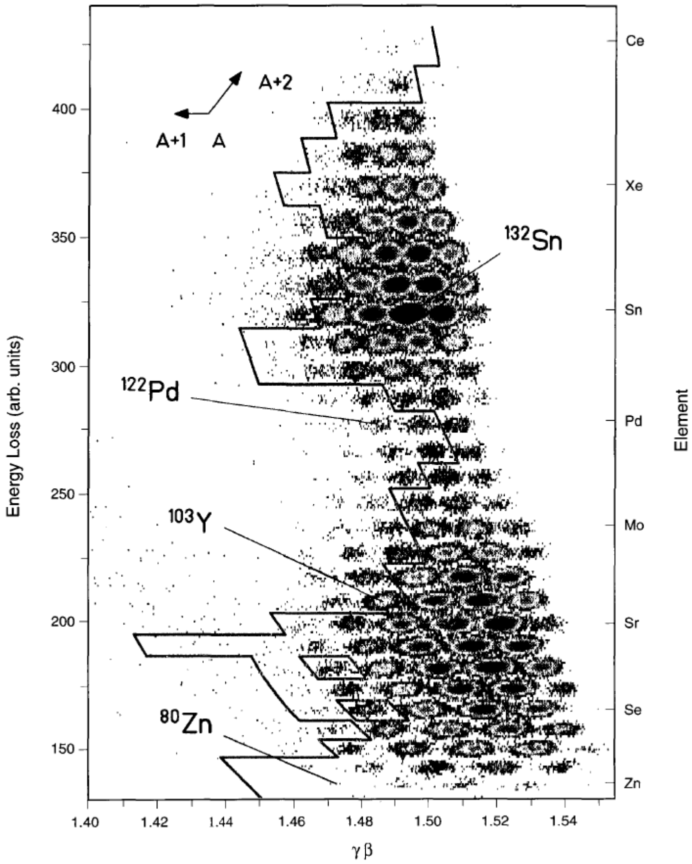
Fission of ^{238}U with high-energy protons at CERN-ISOLDE

H. Ravn, Phys. Rep. 54 (1979) 201

1994: Discovery of ^{86}Ge, ^{88,89}As, ^{90}Se, …

Projectile fission at relativistic velocities: a novel and powerful source of neutron-rich isotopes well suited for in-flight isotopic separation





~60 new isotopes discovered

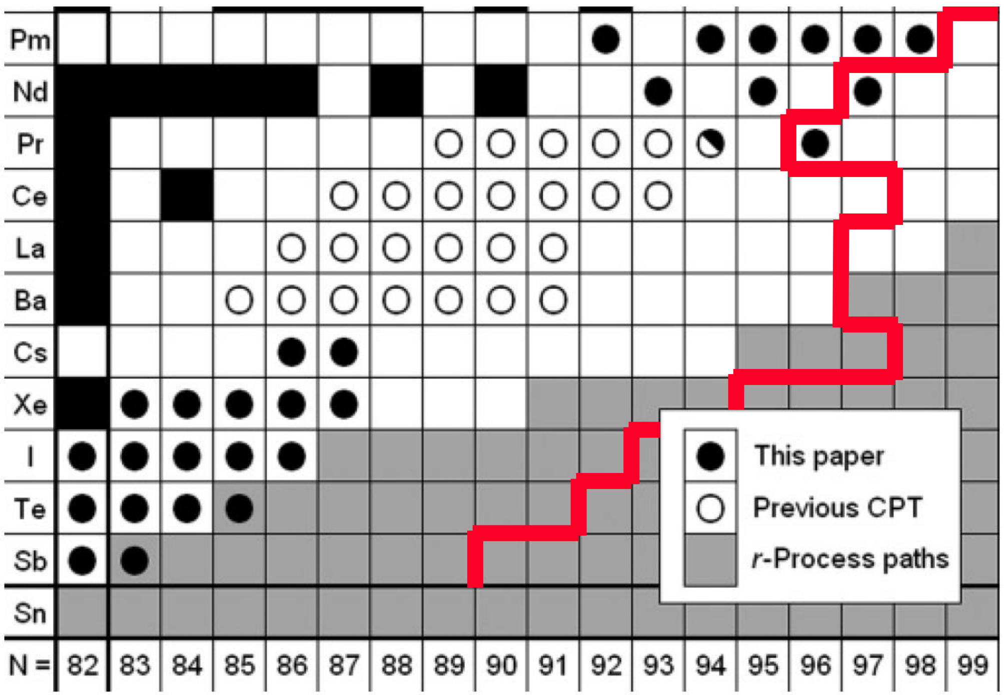
M. Bernas et al., Phys. Lett. B 331 (1994) 19

2012: Discovery of ^{155}Pr and ^{157}Nd

PHYSICAL REVIEW C **85**, 045805 (2012)

Mass measurements near the *r*-process path using the Canadian Penning Trap mass spectrometer

J. Van Schelt,[1,2] D. Lascar,[3,2] G. Savard,[1,2] J. A. Clark,[2] S. Caldwell,[1,2] A. Chaudhuri,[4,2] J. Fallis,[4,2] J. P. Greene,[2]
A. F. Levand,[2] G. Li,[5,2] K. S. Sharma,[4] M. G. Sternberg,[1,2] T. Sun,[2] and B. J. Zabransky[2]



Spontaneous fission of ^{252}Cf at CARIBU

Limit of known isotopes

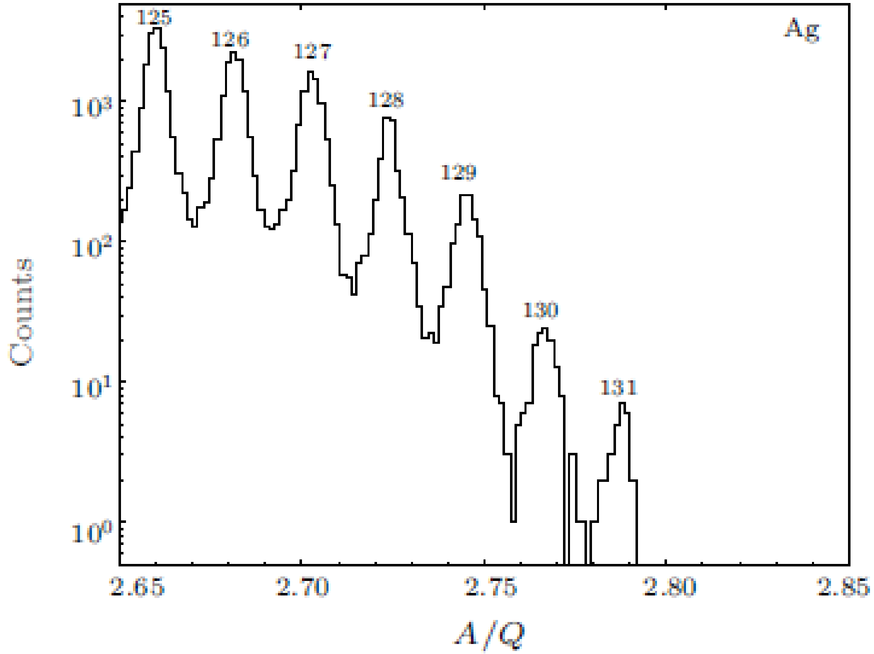
2013: Discovery of ^{131}Ag

CHIN. PHYS. LETT. Vol. 30, No. 4 (2013) 042501

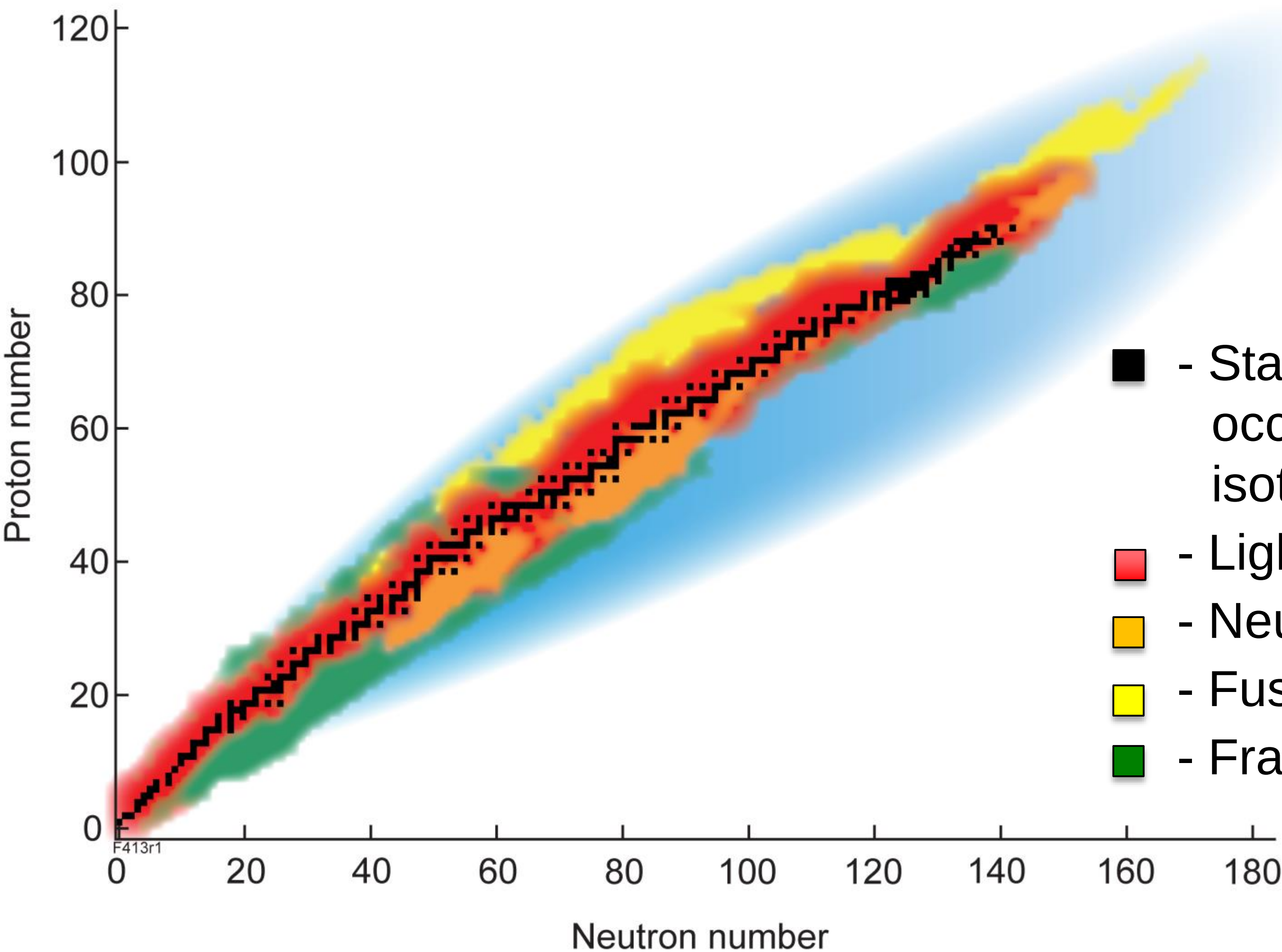
Observation of New Isotope ^{131}Ag via the Two-Step Fragmentation Technique *

WANG He(王赫)[1,2], N. Aoi[3], S. Takeuchi[2], M. Matsushita[4], P. Doornenbal[2], T. Motobayashi[2], D. Steppenbeck[4], K. Yoneda[2], K. Kobayashi[5], J. Lee[2], LIU Hong-Na(刘红娜)[1,2], Y. Kondo[6], R. Yokoyama[4], H. Sakurai[2], YE Yan-Lin(叶沿林)[1]**

We report on the first observation of the neutron-rich nucleus ^{131}Ag. This isotope was produced via fragmentation reactions of intense secondary radioactive ion beams, including ^{134,135}Sn. The secondary beams were produced from induced fission reactions from a stable ^{238}U beam at 345 MeV/nucleon.

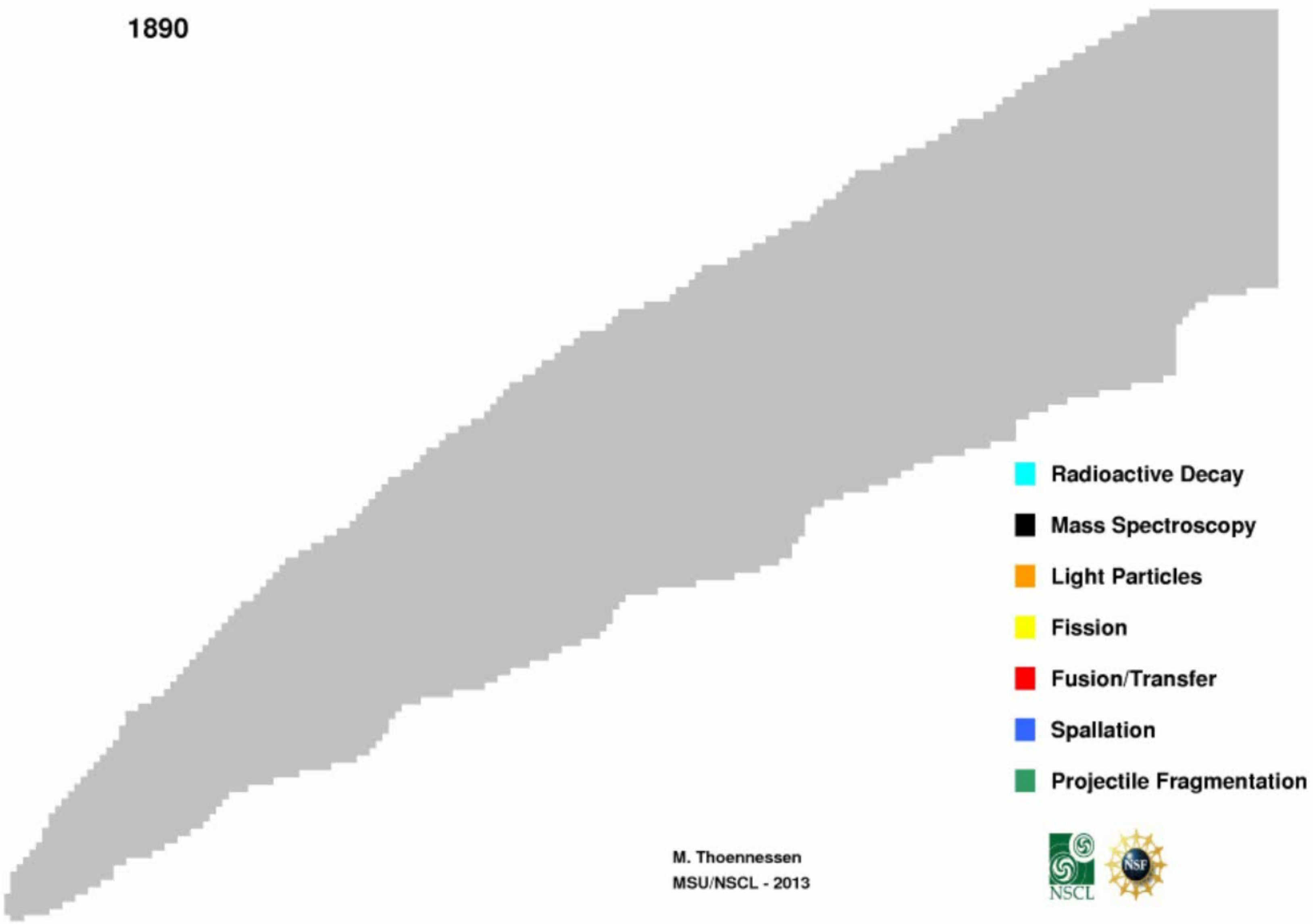


Methods



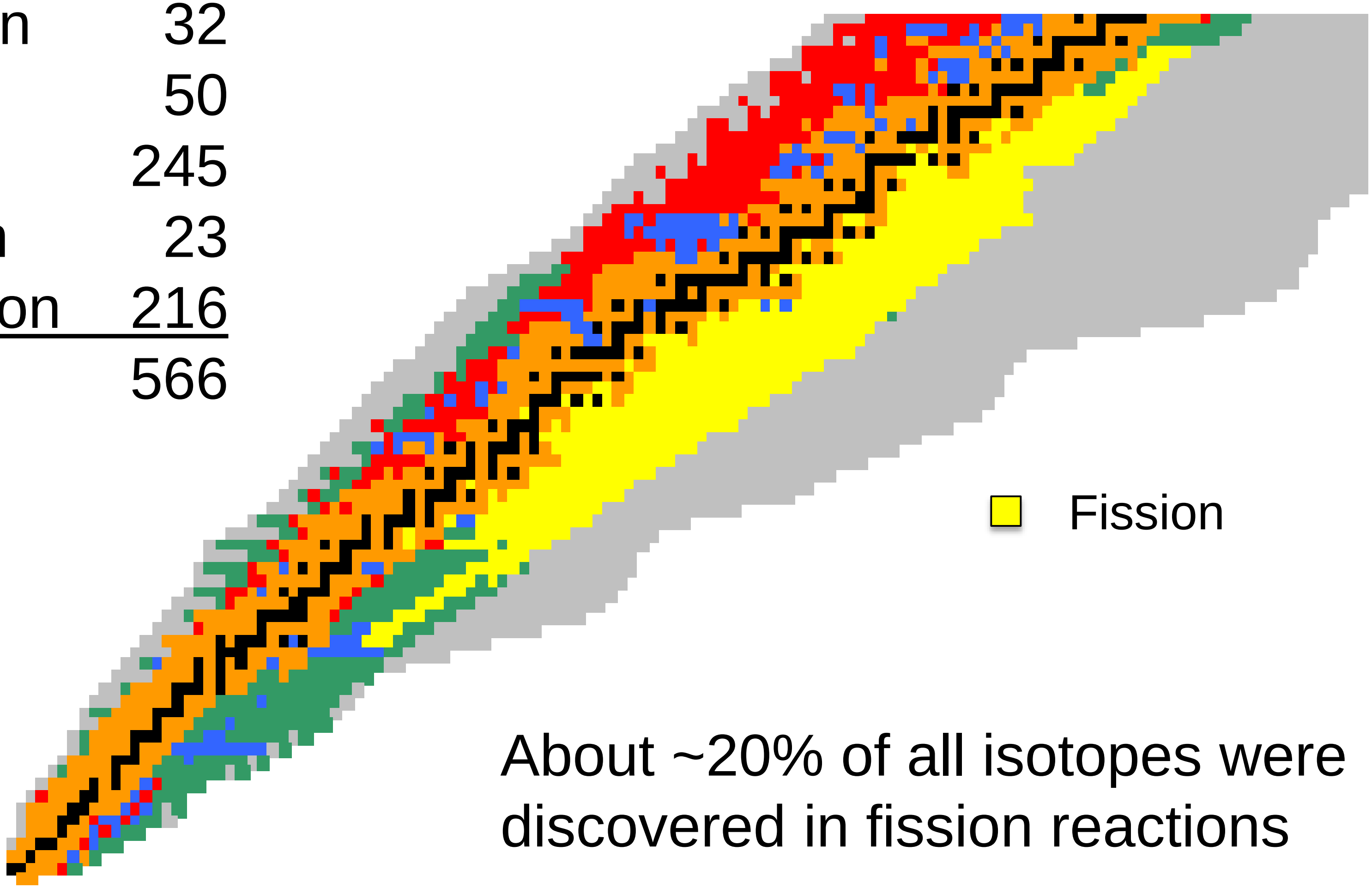
- Stable and naturally occurring radioactive isotopes
- Light-particle reactions
- Neutron reactions
- Fusion-evaporation
- Fragmentation/spallation

http://www.nscl.msu.edu/~thoennes/isotopes



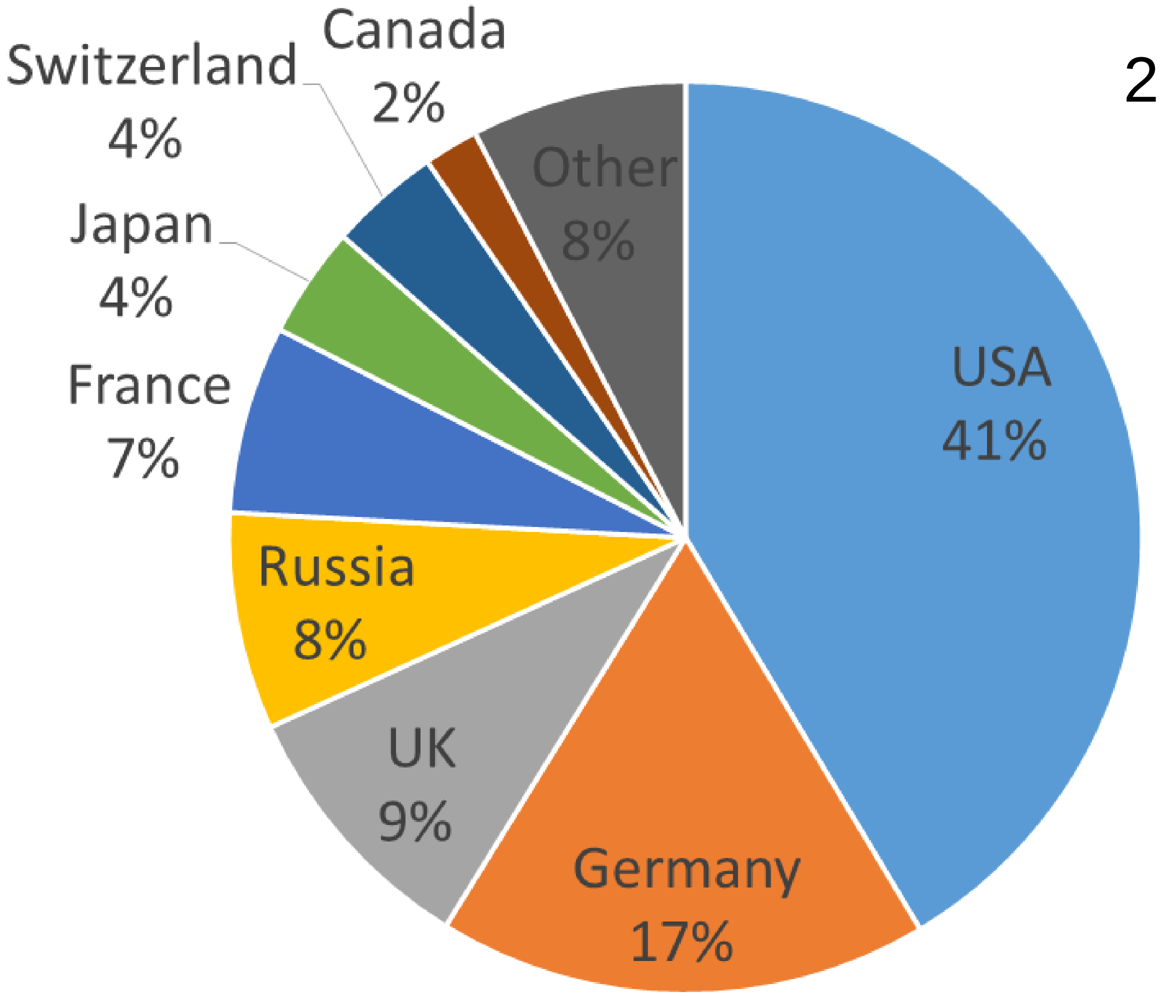
Fission

spont. fission	32
p/d-fission	50
n-fission	245
spall. fission	23
in-flight fission	216
total	566



About ~20% of all isotopes were discovered in fission reactions

Countries



25 different countries

…		
19.	Australia	2
	India	**2**
	Norway	2
22.	Brazil	1
	Hungary	1
	New Zealand	1
	Poland	1

^{191}Ir and ^{193}Ir

SEPTEMBER 14, 1935 NATURE 437

Iridium Isotopes and their Nuclear Spin

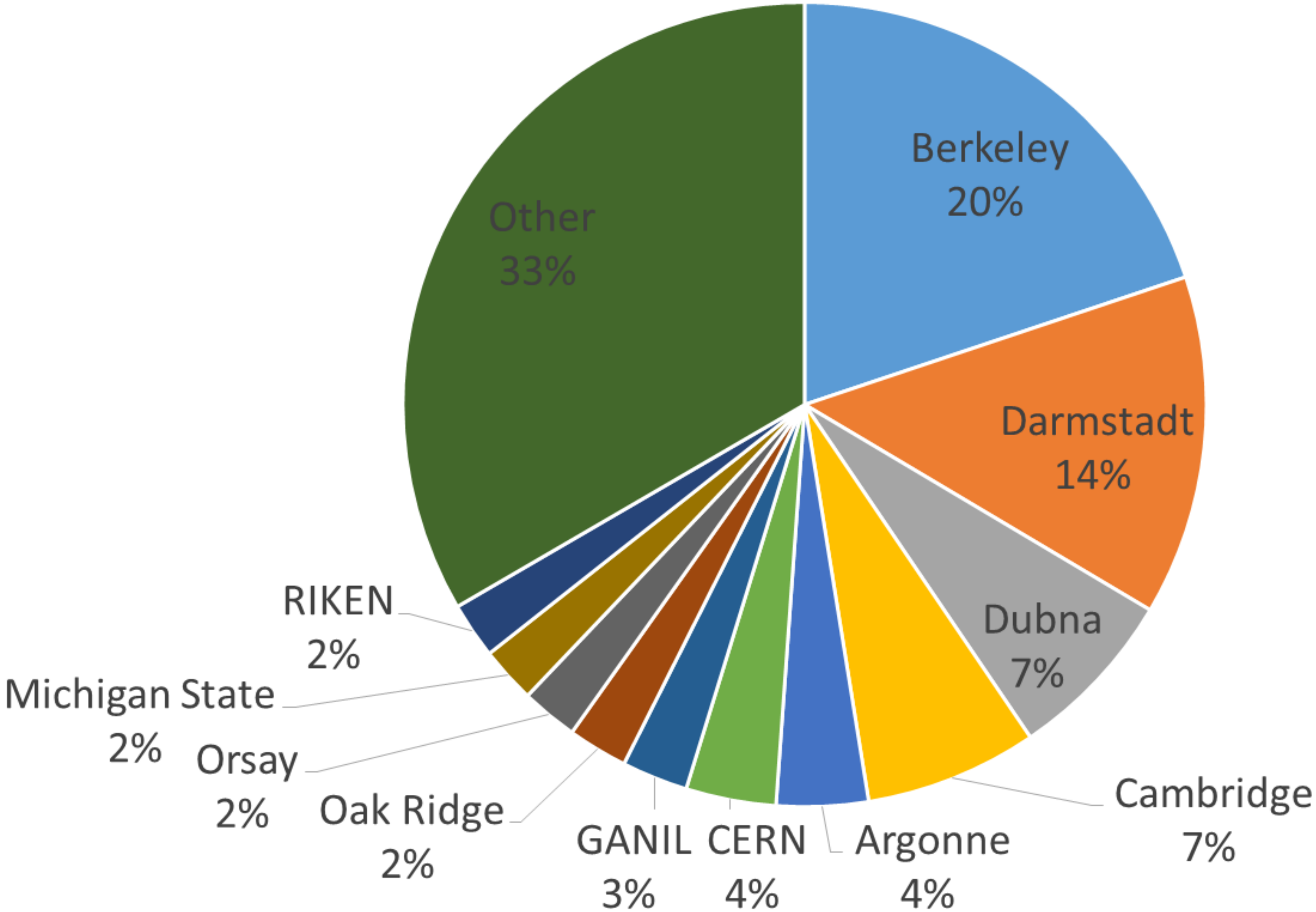
AN examination of the hyperfine structure of the arc lines of iridium λλ3800·10, 3513·67 and 2924·81 A. radiated from a modified form of hollow cathode reveals that the three lines having a common lower level in the ground term $5d^8 6s\ ^4F_{4\frac{1}{2}}$[1] are identical in structure.

The observed structure is accounted for uniquely by assuming two isotopes of masses 191 and 193 with nuclear spins of $\frac{1}{2}$ and $\frac{3}{2}$ respectively.

Department of Physics,
Central College,
Bangalore.
Aug. 7.

B. VENKATESACHAR.
L. SIBAIYA.

Laboratories



Top 10 authors

First Authors:

F. W. Aston	207
M. Bernas	110
J. Kurcewicz	60
Yu. Ts. Oganessian	50
T. Ohnishi	49
H. Alvarez-Pol	39
K. S. Toth	37
S. Hofmann	36
A. J. Dempster	36
D. Guillemaud-Mueller	35

Any Authors:

H. Geissel	273
M. Pfützner	225
G. Münzenberg	218
F. W. Aston	207
P. Armbruster	203
M. Bernas	164
K. Sümmerer	154
A. Heinz	147
T. Kubo	138
D. Bazin	137

Weighted Authors:

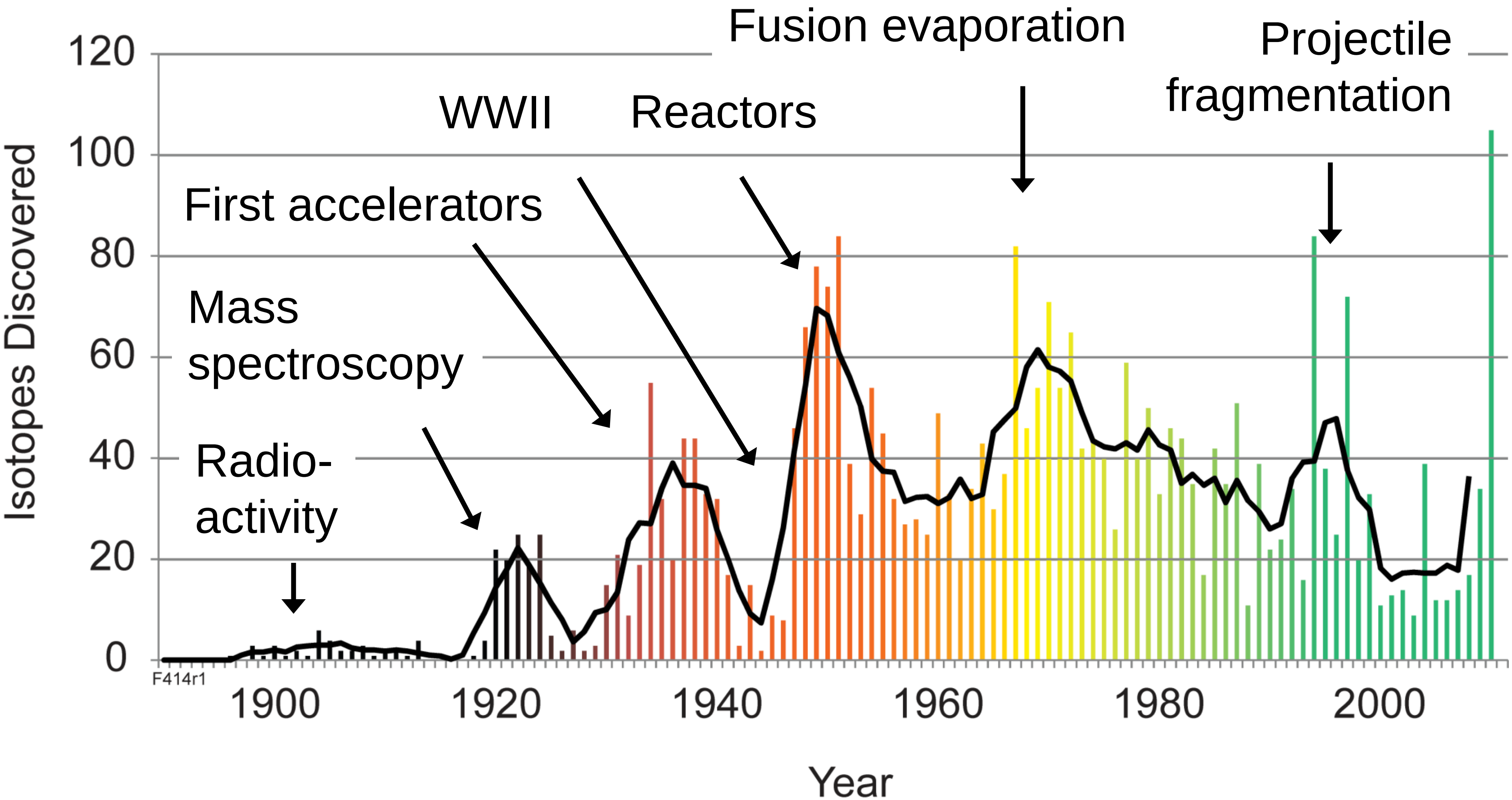
F. W. Aston	206.0
A. J. Dempster	36.0
G. T. Seaborg	33.8
A. Ghiorso	31.2
A. Siivola	25.5
E. K. Hyde	25.3
G. Wilkinson	24.5
M. L. Pool	21.3
R. D. MacFarlane	19.5
P. Armbruster	16.9

Some more random statistics

3518 Different authors
1524 Papers
897 Different first authors

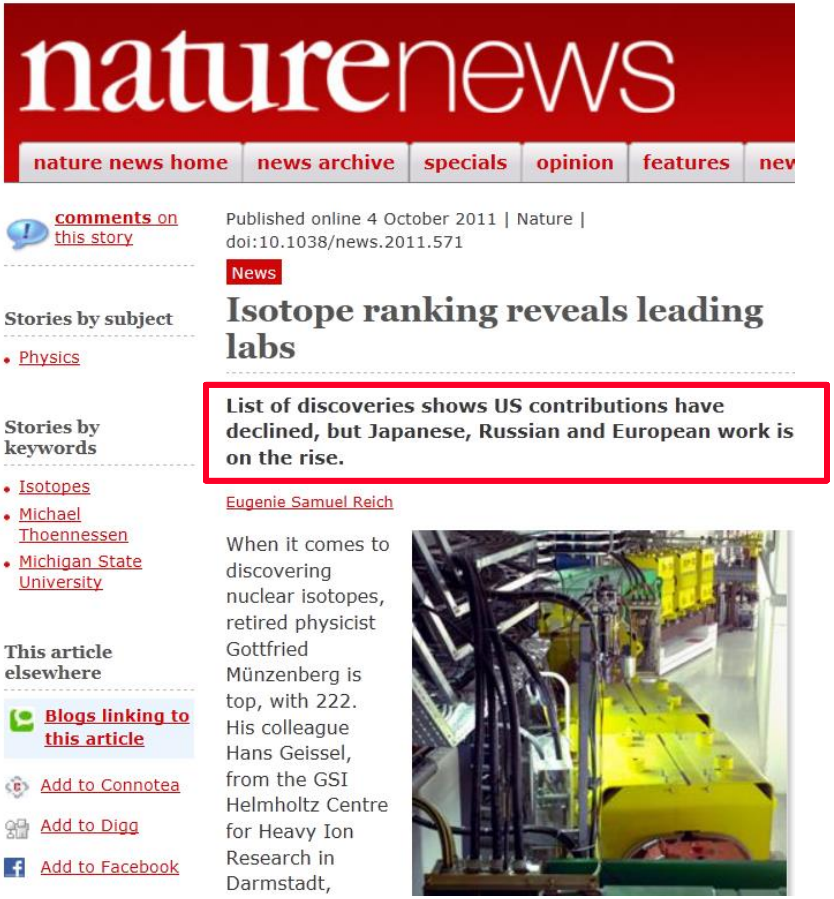
Most first authors:	Aston	207
Most author:	Geissel	273
Most isotopes/paper:	Kurcewicz	60
Most authors/paper:	Khuyagbaatar	70
Most authors/isotope:	Khuyagbaatar	70
Most isotopes/author/paper:	Aston	16

Discoveries per year



M. T. and B.M. Sherrill, Nature 473 (2011) 25

Nature news and trendwatch



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Published online 4 October 2011 | Nature | doi:10.1038/news.2011.571

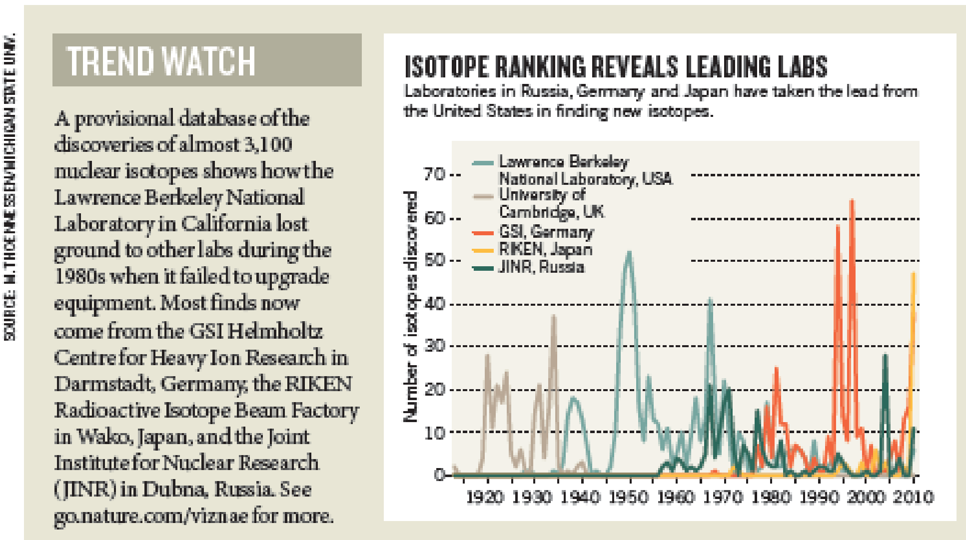
News

Isotope ranking reveals leading labs

List of discoveries shows US contributions have declined, but Japanese, Russian and European work is on the rise.

Eugenie Samuel Reich

When it comes to discovering nuclear isotopes, retired physicist Gottfried Münzenberg is top, with 222. His colleague Hans Geissel, from the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt,



SOURCE: M. THOENNESSEN/MICHIGAN STATE UNIV.

TREND WATCH

A provisional database of the discoveries of almost 3,100 nuclear isotopes shows how the Lawrence Berkeley National Laboratory in California lost ground to other labs during the 1980s when it failed to upgrade equipment. Most finds now come from the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany, the RIKEN Radioactive Isotope Beam Factory in Wako, Japan, and the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. See go.nature.com/viznae for more.

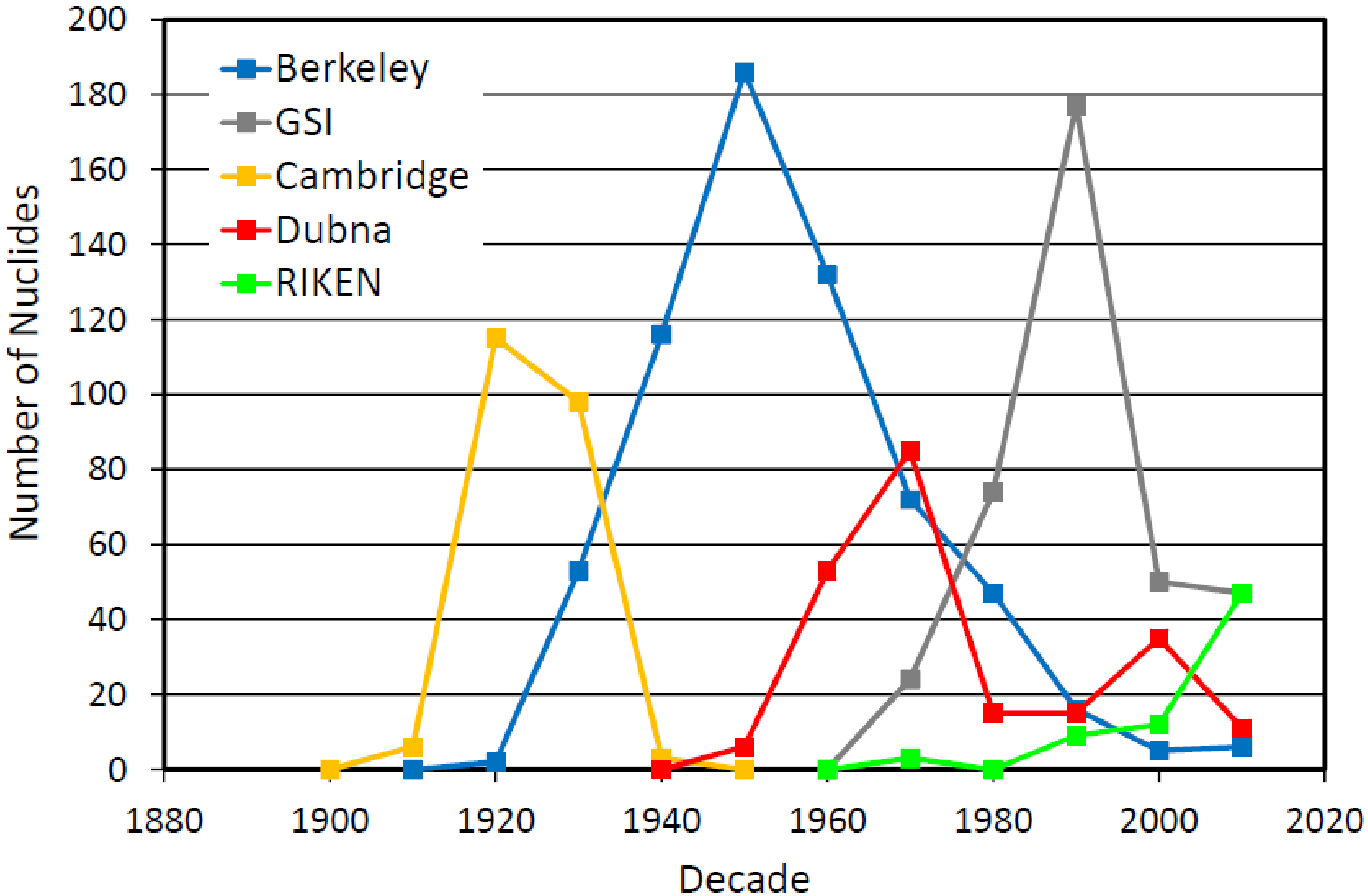
ISOTOPE RANKING REVEALS LEADING LABS

Laboratories in Russia, Germany and Japan have taken the lead from the United States in finding new isotopes.

Nature 478 (2011) 161

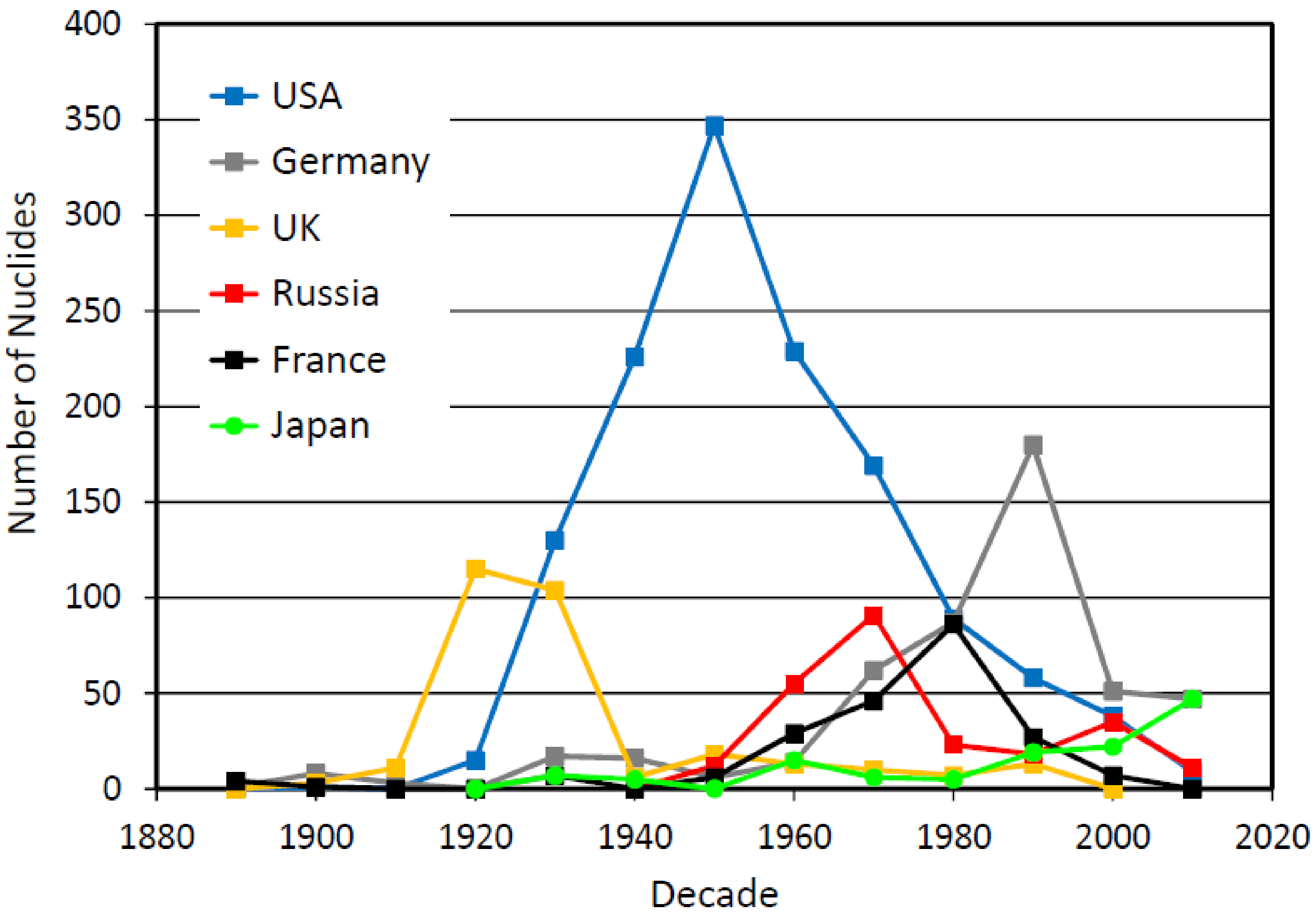
doi:10.1038/news.2011.571

Leading laboratories



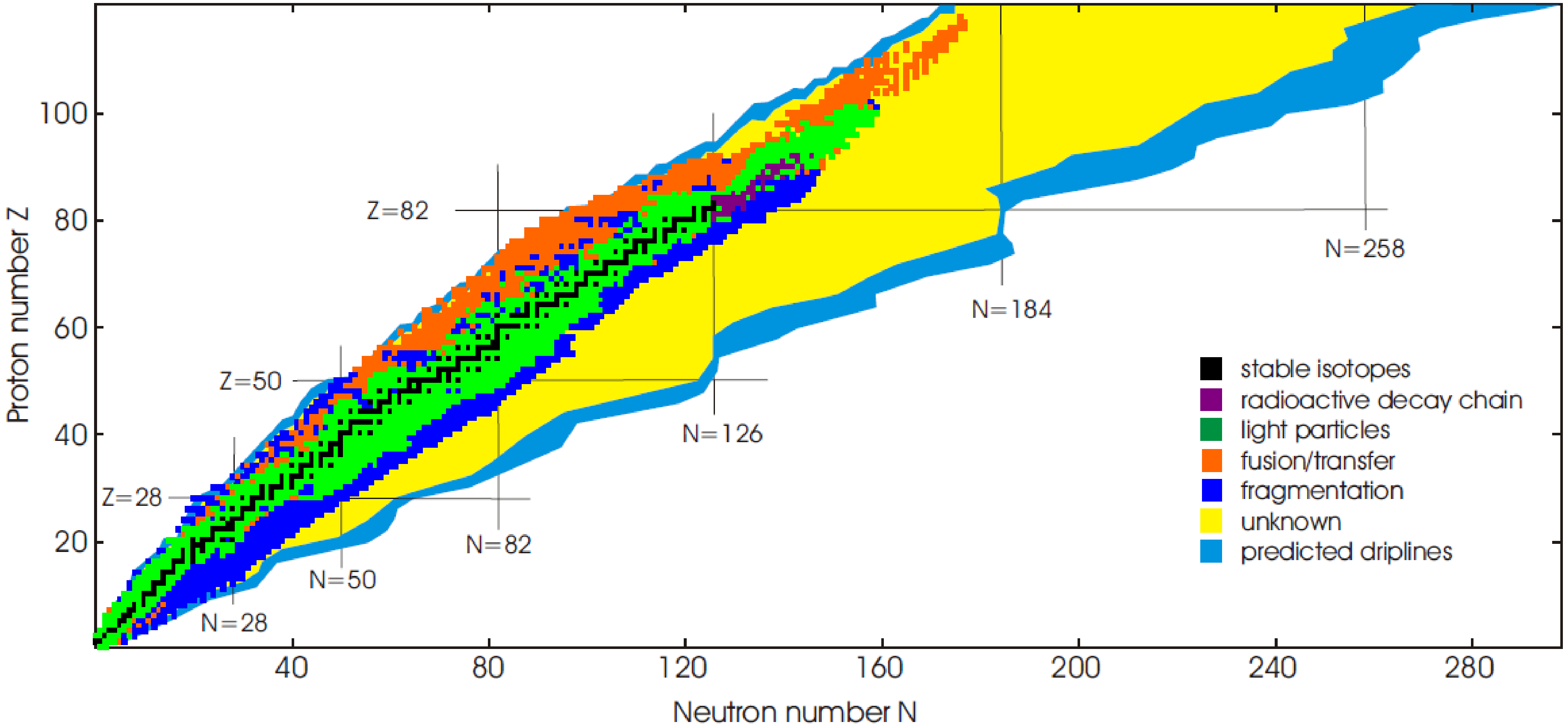
M. T., Nuclear Physics News 22, No.3, 19 (2012)

Leading countries



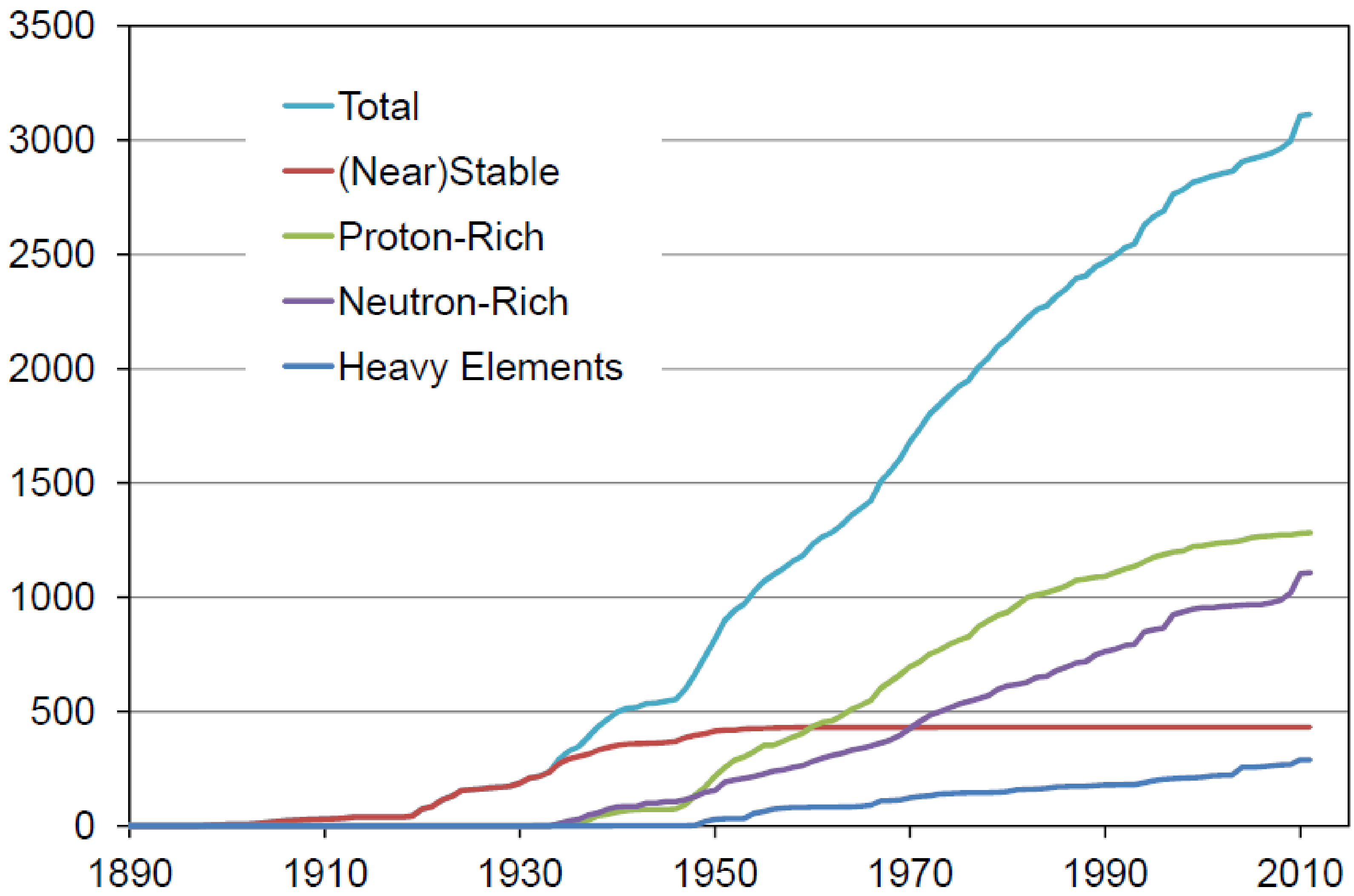
M. T., Nuclear Physics News 22, No.3, 19 (2012)

How many more nuclides are there?

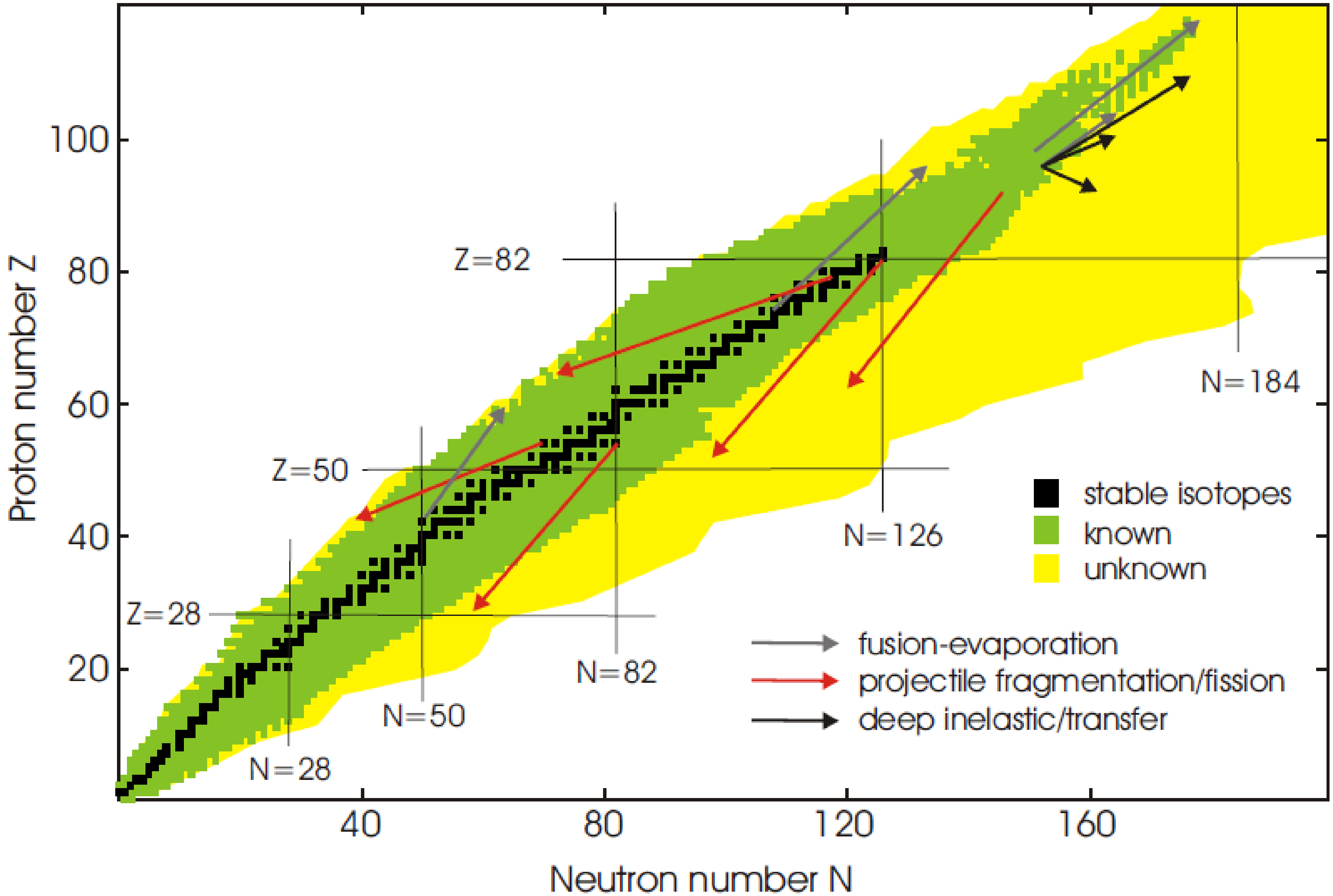


7000 bound nuclide should exist (Erler *et al.*,Nature 486 (2012) 509)

Nuclide discovery



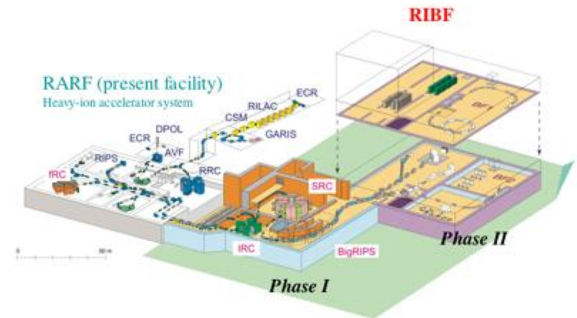
How can new nuclides be discovered?



New fragmentation facilities

RIKEN NiSHiNA CENTER

RIKEN Nishina Center for Accelerator-Based Science

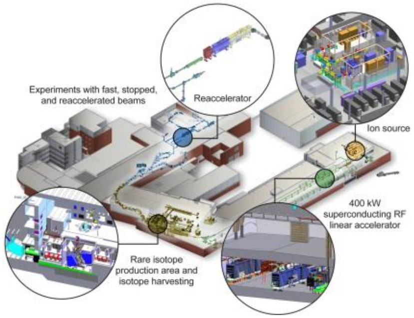


FAIR

FAIR Facility for Antiproton and Ion Research

GSI

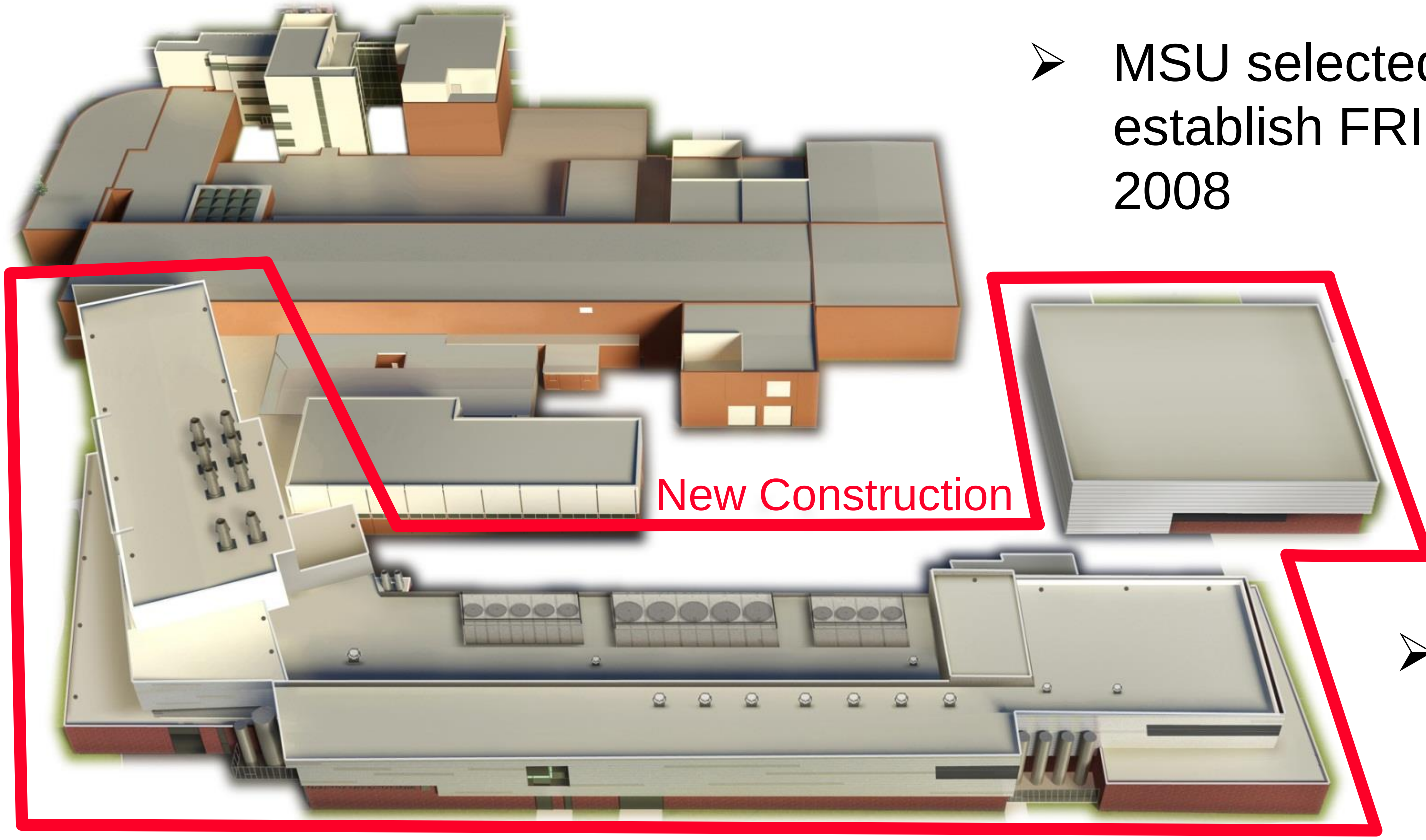
FACILITY FOR RARE ISOTOPE BEAMS



FRIB: Facility for Rare Isotope Beams

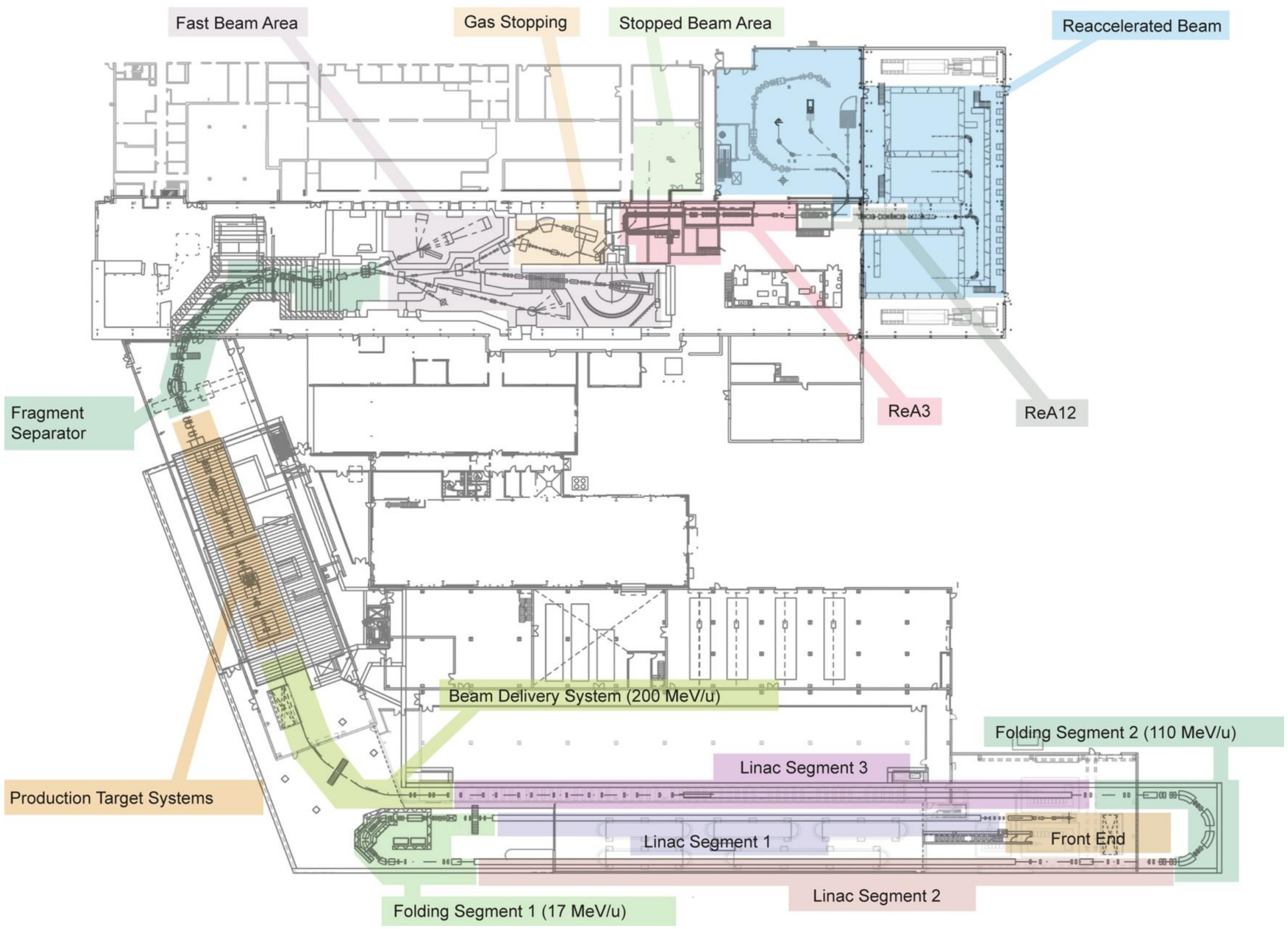
- FRIB is located on the campus of Michigan State University and funded by the U.S. Department of Energy

- MSU selected to design and establish FRIB in December 2008



- Project started in June 2009

New accelerator and present experimental areas



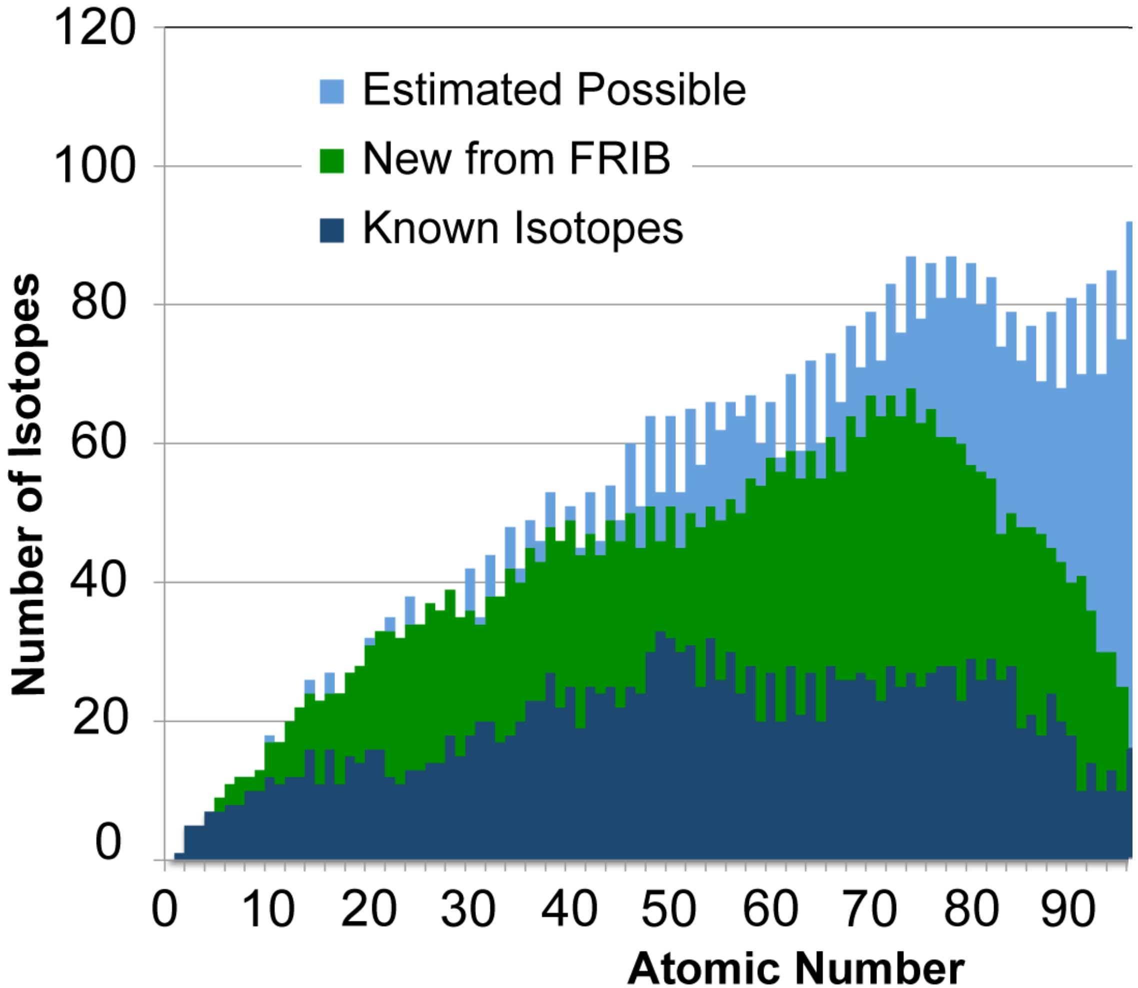
FRIB project is on track

➢ Ground breaking: March 17, 2014
➢ Early completion expected in 2020
➢ Official project completion is 2022

Ground breaking

May 5, 2014

New nuclides with FRIB



~80% of all isotopes for Z<92

Summary

- About 20% of all isotopes were discovered from fission
- Projectile fission is the most promising method to discover a large number of neutron-rich isotopes in the future