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Current status and future potential of nuclide discoveries

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Abstract
Currently about 3000 different nuclei are known with about another 3000–4000 predicted to exist. A review of the discovery of the nuclei, the present status and the possibilities for future discoveries are presented.

(Some figures may appear in colour only in the online journal)

This article was invited by Robert E Tribble.

Contents

1. Introduction

The strong force, responsible for the binding of nucleons, is one of the fundamental forces. In order to understand this force it is critical to know which combination of neutrons and protons can form a bound nuclear system. Even now, after more than 100 years of nuclear physics research this information is only known for the lightest elements. Thus the search for new nuclides with more and more extreme neutron to proton ratios continues to be important. The discovery of new nuclides also is the first step in exploring and measuring any properties of these nuclides.

Over the years more and more sophisticated detectors and powerful accelerators were developed to push the limit of nuclear knowledge further and further. At the present time about 3000 nuclides are known. Recently it was calculated that about 7000 nuclides are bound with respect to neutron or proton emission [1]. In addition, there are neutron and proton-unbound nuclides which can have significantly shorter lifetimes or appear only for a very short time as a resonance. The properties of these nuclides beyond the 'driplines' can also be studied with special techniques [2, 3] and they are especially interesting because they represent the extreme limits for each element.

This review gives a brief historical overview followed by a summary of the present status and a discussion of future perspectives for the discovery of new nuclides. Throughout the paper the word nuclide is used rather than the widely used but technically incorrect term isotope. The term isotope is only appropriate when referring to a nuclide of a specific element.

2. Historical overview

It can be argued that the field of nuclear physics began with the discovery of radioactivity by Becquerel in 1896 [4] who

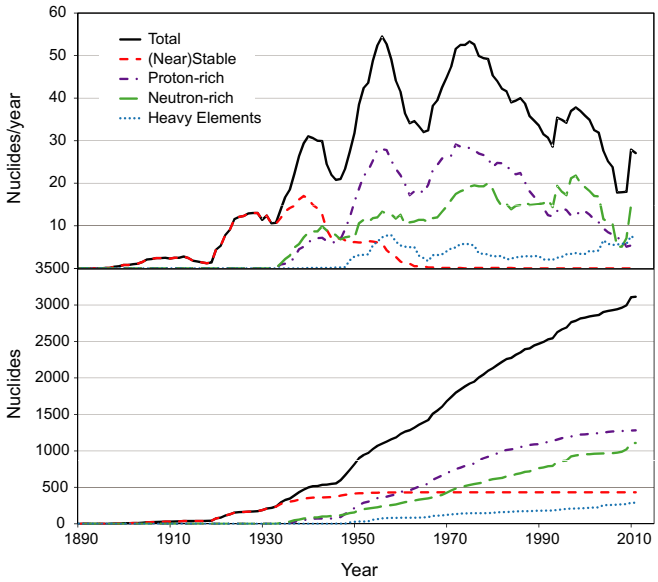

Figure 1. Discovery of nuclides as a function of year. The top figure shows the 10-year running average of the number of nuclides discovered per year while the bottom figure shows the cumulative number. The total number of nuclides shown by the black, solid lines are plotted separately for near-stable (red, short-dashed lines), neutron-deficient (purple, dotted–dashed lines), neutron-rich (green, long-dashed lines) and transuranium (blue, dotted lines) nuclides (see text for explanation).

observed the radioactive decay of what was later determined to be ^{238}U [5, 6]. Subsequently, polonium (^{210}Po [7]) and radium (^{226}Ra [8]) were observed as emitting radioactivity, before Rutherford discovered the radioactive decay law and determined the half-life of radon (^{220}Rn [9]). He was also the first to propose the radioactive decay chains and the connections between the different active substances [10] as well as the identification of the α-particle: '...we may conclude that an α-particle is a helium atom, or, to be more precise, the α-particle, after it has lost its positive charge, is a helium atom' [11].

The distinction of different isotopes for a given element was discovered only in 1913 independently by Fajans [12] and Soddy [13] explaining the relationship of the radioactive chains. Soddy coined the name 'isotope' from the Greek words 'isos' (same) and 'topos' (place) meaning that two different 'isotopes' occupy the same position in the periodic table [14].

The first clear identification of two isotopes of an element other than in the radioactive decay chains was reported by Thomson in 1913 using the positive-ray method: 'There can, therefore, I think, be little doubt that what has been called neon is not a simple gas but a mixture of two gases, one of which has an atomic weight about 20 and the other about 22' [15].

Since this first step, continuous innovations of new experimental techniques utilizing the new knowledge gained about nuclides led to the discovery of additional nuclides. This drive to discover more and more exotic nuclides has moved the field forward up to the present day. Figure 1 demonstrates this development where the number of nuclides discovered per year (top) and the integral number of discovered nuclides (bottom) are shown. In addition to the total number of nuclides (black, solid lines) the figure also shows the number of near-stable (red, short-dashed lines), neutron-deficient (purple, dotted–dashed lines), neutron-rich (green, long-dashed lines) and transuranium (blue, dotted lines) nuclides. Near-stable nuclides are all nuclides between the most neutron-deficient and neutron-rich stable isotopes of a given element. Lighter and heavier radioactive isotopes of the elements are then classified as neutron-deficient and neutron-rich, respectively.

The figure shows that the rate of discovery was not smooth and the peaks can be directly related to the development of new experimental techniques as explained in the next subsections.

2.1. Mass spectroscopy of stable nuclides

In 1908, Rutherford and Geiger had identified the α-particle as helium [11] and in 1913 Thompson accepted in addition to neon with mass number 20 the presence of a separate neon substance with mass number 22 which represented the beginning of mass spectroscopic methods to identify isotopes as separate identities of the same element with different mass numbers [15]. The first 'mass spectra' were measured by Aston when he added focusing elements to his first 'positive ray spectrograph' in 1919 [16]. From 1919 to 1930 the number of known identified nuclides jumped from 40 to about 200 mostly due to Aston's work. The development of more sophisticated mass spectrographs by Aston [17, 18] and others [19–21] led to the discovery of essentially most of the stable nuclides [22].

2.2. Nuclear reactions and first accelerators

In 1919 Rutherford discovered nuclear transmutation: 'from the results so far obtained it is difficult to avoid the conclusion that the long-range atoms arising from collision of α particles with nitrogen are not nitrogen atoms but probably atoms of hydrogen, or atoms of mass 2' [23]. He apparently observed the reaction ^{14}N(α,p); however, it took six years before Blackett identified the reaction residue as the new nuclide ^{17}O [24]. It took another seven years before in 1932 the discovery of the neutron by Chatwick [25] and the first successful construction of a particle accelerator by Cockcroft and Walton [26] led to the production of many new nuclides by nuclear reactions.

Cockcroft and Walton were able to prove the production of ^{8}Be using their accelerator: '...the lithium isotope of mass 7 occasionally captures a proton and the resulting nucleus of mass 8 breaks into two α-particles...' [27]; Harkins, Gans and Newson produced the first new nuclide (^{16}N) induced by neutrons (^{19}F(n,α)) [28] and in 1934, Curie and Joliot observed artificially produced radioactivity (^{13}N and ^{30}P)[1] in (α,n) reactions for the first time [29].

Also in 1934, Fermi claimed the discovery of a transuranium element in the neutron bombardment of uranium [30]. Although the possibility of fission was immediately mentioned by Noddack: 'It is conceivable that [...] these nuclei decay into several larger pieces' [31], even with mounting evidence in further experiments, Meitner, Hahn and

[1] They also reported another activity assigned to ^{27}Si; however, most likely they observed ^{28}Al.

Strassmann did not take this step: 'These results are hard to understand within the current understanding of nuclei' [32] and 'As chemists 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' [33]. After Meitner and Frisch correctly interpreted the data as fission in 1939 [34], Hahn and Strassmann identified ^{140}Ba [35] in the neutron induced fission of uranium. The first transuranium nuclide (^{239}Np) was then discovered a year later by McMillan and Abelson in neutron capture reactions on ^{238}U [36].

Light-particle induced reactions using α-sources, neutron irradiation, fission, and continuously improved particle accelerators expanded the chart of nuclei toward more neutron-deficient, neutron-rich and further transuranium nuclides for the next two decades. The number of nuclides produced every year continued to increase only interrupted by World War II. By 1950 the existing methods had reached their limits and the number of new isotopes began to drop. New technical developments were necessary to reach isotopes further removed from stability.

2.3. Heavy-ion fusion–evaporation reactions

Although Alvarez demonstrated already in 1940 that it was possible to accelerate ions heavier than helium in the Berkeley 37-inch cyclotron [37], the next major breakthrough came in 1950 when Miller *et al* successfully accelerated detectable intensities of completely stripped carbon nuclei in the Berkeley 60-inch cyclotron [38]. Less than two months later Ghiorso *et al* reported the discovery of ^{246}Cf in the heavy-ion fusion–evaporation reaction ^{238}U(^{12}C,4n) [39]. This represented the first correct identification of a californium nuclide because the discovery of the element californium claimed the observation of ^{244}Cf [40] which was later reassigned to ^{245}Cf [41].

With continuous increases of beam energies and intensities fusion–evaporation reactions became the dominant tool to populate and study neutron-deficient nuclei. The peak in the overall production rate of new nuclides around 1960 is predominantly due to the production of new neutron-deficient nuclides and new superheavy elements. Fusion–evaporation reactions are currently still the only way to produce superheavy elements. The discovery of new elements relies on even further improvements in beam intensities and innovations in detector technology.

2.4. Target and projectile fragmentation

The significant beam energy increases of light-ion as well as heavy-ion accelerators opened up new ways to expand the nuclear chart. In the spallation or fragmentation of a uranium target bombarded with 5.3 GeV protons, Poskanzer *et al* were able to identify several new neutron-rich light isotopes for the first time (^{11}Li, ^{12}Be and ^{14,15}B) in 1966 [42]. Target fragmentation reactions were effectively utilized to produce new neutron-rich nuclides (see, for example, [43]) using the isotope separation on-line (ISOL) method. This technique was developed already 15 years earlier for fission of uranium by Kofoed-Hansen and Nielsen who discovered ^{90}Kr and ^{90,91}Rb [44] .

The inverse reaction, the fragmentation of heavy projectiles on light-mass targets was successfully applied to produce new nuclides for the first time in 1979 by bombarding a beryllium target with 205 MeV/nucleon ^{40}Ar ions [45]. Projectile fragmentation began to dominate the production of especially neutron-rich nuclei starting in the late 1980s when dedicated fragment separators came online. For an overview of the various facilities, for example, the LISE3 spectrometer at GANIL [46], the RIPS separator at RIKEN [47], the A1200 and A1900 separators at NSCL [48, 49], and the FRS device at GSI [50] see [51]. In addition to these separators a significant number of nuclides were discovered at storage rings, see, for example, [52, 53].

The most recent increase in the production rate of new nuclides is predominantly due to new technical advances at GSI [53–55] and the new next generation radioactive beam facility RIBF [56] with the separator BigRIPS [57] at RIKEN.

2.5. Discoveries of isotopes, isotones, and isobars

It is interesting to follow the discovery of nuclides over the years as a function of isotopes (Z = constant), isotones (N = constant) and isobars (A = constant) as shown in the top, middle and bottom panels of figure 2, respectively.

Unique characteristics of isotopes of elements from the radioactive decay chains were determined around 1900, and although the concept of isotopes was not established at that time these observations can be taken as the first identification of isotopes of these elements. For most of the elements up to $Z = 60$ the first isotope was discovered in the early 1920s with exception of the transition metals of the fifth period between niobium and palladium which were identified for the first time in the 1930s. Also, as mentioned earlier, isotopes of helium (^{4}He or the α-particle [11]) and neon (^{20,22}Ne [15]) were discovered earlier and the neutron was discovered in 1932 [25].

Isotopes of the remaining stable elements were identified by the late 1930s. The last four missing elements below uranium were discovered by the identification of their specific isotopes. They were technetium ($Z = 43$) in 1938 [58], francium ($Z = 87$) in 1939 [59], astatine ($Z = 85$) in 1940 [60], and promethium ($Z = 61$) in 1947 [61]. Transuranium elements were then discovered starting in 1940 with the identification of neptunium (^{239}Np) [36] at an approximately constant rate of about one element every three years (also see figure 5).

Plotting the year of discovery as a function of isotones reveals another pattern. In the light-mass region—approximately between chlorine and zirconium ($N \sim 20$–50)—the even-N isotones were discovered around 1920 while it took about another 15 years before the odd-N isotones were identified. This is due to the significantly smaller abundances of the even-Z/odd-N isotones in this mass region. In contrast, the abundances are more equally distributed in the lanthanide region ($N \sim 80$–110). While the advances in the discovery of new elements were fairly constant, the discovery of isotones displays a different pattern.

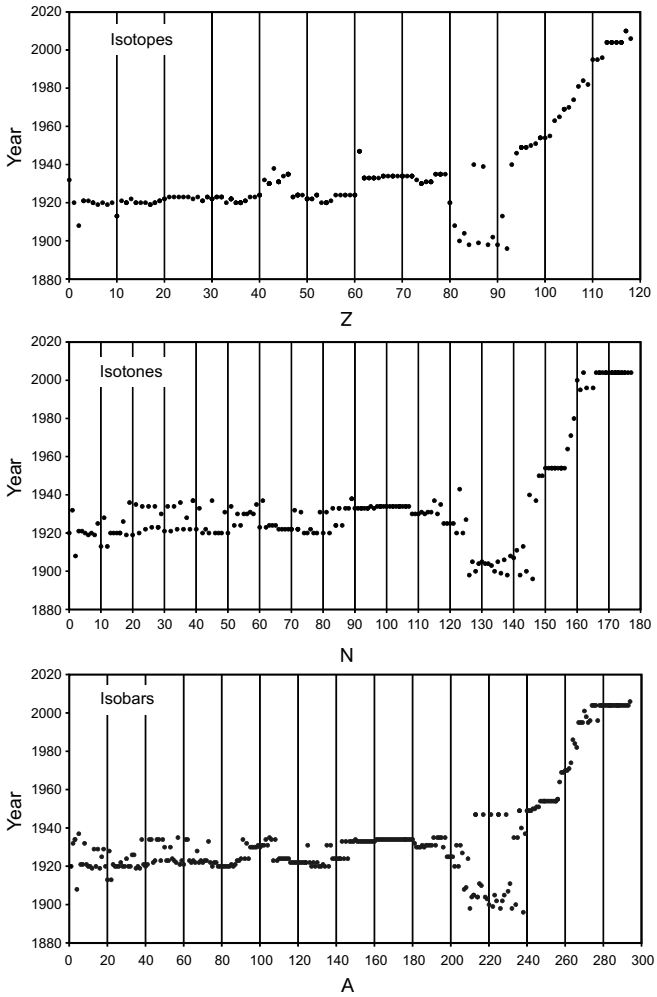

Figure 2. Discoveries of isotopes (top), isotones (middle) and isobars (bottom).

Although intense neutron irradiation of plutonium in the Idaho Materials Test Reactor did not discover any new elements, the successive neutron capture reactions produced many new isotones. In 1954 alone seven new isotones (N = 150–156) were discovered. However, in the following 40 years only one additional isotone was added per decade.

At Dubna hot fusion reactions were used to populate new elements leading to the discovery of 15 new isotones within one year (2004) up to the heaviest currently known isotone of $N = 177$. The recent discovery of element 117 and 118 did not push the isotone limit any further. It should be mentioned that the isotone $N = 164$ has not yet been identified (see also section 4.4).

The pattern of the discovery as a function of mass number up to $A \sim 200$ shown in the bottom panel of figure 2 mirrors approximately the pattern of the isotones. Until 1937, when Meitner *et al* [32] discovered ^{239}U, the discovery of radioactivity by Becquerel in 1896 [4] later attributed to ^{238}U represented the heaviest nuclide. The missing (4n + 3) radioactive decay chain observed in 1943 by Hagemann *et al* [62] filled in the gaps at masses 213, 217, 221, 225 and 229. Currently the heaviest element ($Z = 118$) also represents the heaviest nuclide ($A = 294$).

3. Current status

Recently a comprehensive overview of the discovery of all nuclides was completed [63]. Details of the discovery of 3067 nuclides were described in a series of papers beginning in 2009 [64] with the latest ones being currently published. During this time another 38 nuclides were discovered for a total of 3105 nuclides observed by the end of 2011. Table 1 lists the total number and the range of currently known isotopes for each element. It should be mentioned that for some elements not all isotopes between the most neutron-deficient and the most neutron-rich isotopes have been observed. In light neutron-rich nuclei these are ^{21}C, ^{30}F, ^{33}Ne, ^{36}Na and ^{39}Mg. The cases in the neutron-deficient medium-mass and the superheavy mass region are discussed in sections 4.2 and 4.4, respectively. The table also lists the year of the first and most recent discovery as well as the reference for the detailed documentation of the discovery.

While the recognition for the discovery of a new element is well established with strict criteria set by the International Union of Pure and Applied Chemistry (IUPAC) and the International Union of Pure and Applied Physics (IUPAP) [65, 66] the discovery of the different isotopes for a given element is not well defined [67]. The nuclides included in table 1 had to be (1) clearly identified, either through decay-curves and relationships to other known nuclides, particle or γ-ray spectra, or unique mass and element identification, and (2) published in a refereed journal. In order to avoid setting an arbitrary lifetime limit for the definition of the existence of a nuclide, particle-unbound nuclides with only short-lived resonance states were included. Isomers were not considered separate nuclides.

The element with the most isotopes (46) currently known is mercury, followed by thallium, lead and polonium with 42 each. The element with the fewest isotopes is element 118 where only one isotope ($A = 294$) is currently known. The heaviest nuclides are ^{294}117 and ^{294}118. However, it should be stressed that the observation of elements 117 and 118 has not been accepted by IUPAC.

4. Potential discoveries in the near future

The 3015 nuclides currently reported in the published literature still probably constitute less than 50% of all nuclides that potentially could be observed. In the following subsections nuclides which should be discovered in the near future are discussed.

4.1. Proceedings and internal reports

Until the end of 2011 twenty-six nuclides had only been reported in conference proceedings or internal reports. Table 2 lists these nuclides along with the author, year, laboratory, conference or report and reference of the discovery. Most of them were reported at least ten years ago, so that it is unlikely that these results will be published in refereed journals in the future. Conference proceedings quite often contain preliminary results and it is conceivable that these results then do not hold up for a refereed journal.

Table 1. Discovery of the isotopes of all elements. The total number of isotopes, lightest and heaviest isotope and the year of first and most recent discovery is listed. The last column refers to the publication where the details of the discoveries are compiled.

Element	Z	No of Iso.	Lightest	Heaviest	First	Last	Ref.
Neutron(s)	0	2	1	2	1932	1965	[63]
Hydrogen	1	7	1	7	1920	2003	[63]
Helium	2	9	2	10	1908	1994	[63]
Lithium	3	10	4	13	1921	2008	[63]
Beryllium	4	9	6	14	1921	1983	[63]
Boron	5	13	7	19	1920	2010	[63]
Carbon	6	14	8	22	1919	1986	[63]
Nitrogen	7	14	10	23	1920	2002	[63]
Oxygen	8	14	12	25	1919	2008	[63]
Fluorine	9	16	14	31	1920	2010	[63]
Neon	10	18	16	34	1913	2002	[63]
Sodium	11	19	18	37	1921	2004	[68]
Magnesium	12	21	19	40	1920	2007	[68]
Aluminum	13	22	22	43	1922	2007	[68]
Silicon	14	23	22	44	1920	2007	[68]
Phosphorus	15	21	26	46	1920	1990	[68]
Sulfur	16	22	27	48	1920	1990	[68]
Chlorine	17	21	31	51	1919	2009	[68]
Argon	18	23	31	53	1920	2009	[68]
Potassium	19	22	35	56	1921	2009	[68]
Calcium	20	24	35	58	1922	2009	[69]
Scandium	21	23	39	61	1923	2009	[70]
Titanium	22	25	39	63	1923	2009	[70]
Vanadium	23	24	43	66	1923	2009	[71]
Chromium	24	27	42	68	1923	2009	[72]
Manganese	25	26	46	71	1923	2010	[72]
Iron	26	30	45	74	1922	2010	[73]
Cobalt	27	27	50	76	1923	2010	[74]
Nickel	28	32	48	79	1921	2010	[72]
Copper	29	28	55	82	1923	2010	[72]
Zinc	30	32	54	85	1922	2010	[75]
Gallium	31	28	60	87	1923	2010	[76]
Germanium	32	31	60	90	1923	2010	[76]
Arsenic	33	29	64	92	1920	1997	[77]
Selenium	34	32	64	95	1922	2010	[75]
Bromine	35	30	69	98	1920	2011	[75]
Krypton	36	33	69	101	1920	2010	[78]
Rubidium	37	31	73	103	1921	2010	[79]
Strontium	38	35	73	107	1923	2010	[79]
Yttrium	39	34	76	109	1923	2010	[80]
Zirconium	40	35	78	112	1924	2010	[80]
Niobium	41	34	82	115	1932	2010	[80]
Molybdenum	42	35	83	117	1930	2010	[79]
Technetium	43	35	86	120	1938	2010	[80]
Ruthenium	44	38	87	124	1931	2010	[80]
Rhodium	45	38	89	126	1934	2010	[79]
Palladium	46	38	91	128	1935	2010	[81]
Silver	47	38	93	130	1923	2000	[82]
Cadmium	48	38	96	133	1924	2010	[83]
Indium	49	38	98	135	1924	2002	[69]
Tin	50	39	100	138	1922	2010	[69]
Antimony	51	38	103	140	1922	2010	[81]
Tellurium	52	39	105	143	1924	2010	[81]
Iodine	53	38	108	145	1920	2010	[81]
Xenon	54	40	109	148	1920	2010	[81]
Cesium	55	41	112	152	1921	1994	[84]
Barium	56	39	114	152	1924	2010	[85]
Lanthanum	57	35	117	153	1924	2001	[84]
Cerium	58	35	121	155	1924	2005	[64]
Praseodymium	59	32	121	154	1924	2005	[84]
Neodymium	60	31	125	156	1924	1999	[75]
Promethium	61	32	128	159	1947	2005	[84]
Samarium	62	34	129	162	1933	2005	[86]
Europium	63	35	130	166	1933	2008	[86]
Gadolinium	64	31	135	166	1933	2005	[86]
Terbium	65	31	135	168	1933	2004	[86]
Dysprosium	66	32	139	170	1934	2010	[87]
Holmium	67	32	140	172	1934	2001	[87]
Erbium	68	32	144	175	1934	2003	[87]
Thulium	69	33	145	177	1934	1998	[87]
Ytterbium	70	31	149	180	1934	2001	[87]
Lutetium	71	35	150	184	1934	1993	[76]
Hafnium	72	36	154	189	1934	2009	[76]
Tantalum	73	38	155	192	1932	2009	[88]
Tungsten	74	38	157	194	1930	2010	[89]
Rhenium	75	39	159	197	1931	2011	[88]
Osmium	76	41	161	201	1931	2011	[88]
Iridium	77	40	165	204	1935	2011	[88]
Platinum	78	40	166	205	1935	2010	[69]
Gold	79	41	170	210	1935	2011	[90]
Mercury	80	46	171	216	1920	2010	[70]
Thallium	81	42	176	217	1908	2010	[91]
Lead	82	42	179	220	1900	2010	[91]
Bismuth	83	41	184	224	1904	2010	[91]
Polonium	84	42	186	227	1898	2010	[91]
Astatine	85	39	191	229	1940	2010	[92]
Radon	86	39	193	231	1899	2010	[92]
Francium	87	35	199	233	1939	2010	[92]
Radium	88	34	201	234	1898	2005	[92]
Actinium	89	31	206	236	1902	2010	[93]
Thorium	90	31	208	238	1898	2010	[93]
Protactinium	91	28	212	239	1913	2005	[93]
Uranium	92	23	217	242	1896	2000	[93]
Neptunium	93	20	225	244	1940	1994	[94]
Plutonium	94	20	228	247	1946	1999	[94]
Americium	95	16	232	247	1949	2000	[94]
Curium	96	17	233	251	1949	2010	[94]
Berkelium	97	13	238	251	1950	2003	[94]
Californium	98	20	237	256	1951	1995	[94]
Einsteinium	99	17	241	257	1954	1996	[70]
Fermium	100	19	241	259	1954	2008	[95]
Mendelevium	101	16	245	260	1955	1996	[95]
Nobelium	102	11	250	260	1963	2001	[95]
Lawrencium	103	9	252	260	1965	2001	[95]
Rutherfordium	104	13	253	267	1969	2010	[95]
Dubnium	105	11	256	270	1970	2010	[95]
Seaborgium	106	12	258	271	1974	2010	[95]
Bohrium	107	10	260	274	1981	2010	[95]
Hassium	108	12	263	277	1984	2010	[95]
Meitnerium	109	7	266	278	1982	2010	[95]
Darmstadtium	110	8	267	281	1995	2010	[95]
Roengtenium	111	7	272	282	1995	2010	[95]
Copernicium	112	6	277	285	1996	2010	[95]
113	113	6	278	286	2004	2010	[95]
Flerovium	114	5	285	289	2004	2010	[95]
115	115	4	287	290	2004	2010	[95]
Livermorium	116	4	290	293	2004	2004	[95]
117	117	2	293	294	2010	2010	[95]
118	118	1	294	294	2006	2006	[95]

A curious case is the reported discovery of ^{155,156}Pr and ^{157,158}Nd in the proceedings of RNB-3 in 1996 [96] where these nuclides were included as newly discovered in a figure of the chart of nuclides. The authors also stated: 'In this first experiment, 54 new isotopes were discovered, ranging from $^{86}_{32}$Ge to $^{158}_{60}$Nd' [96]. However, in the original publication only 50 new isotopes were listed and there was no evidence for the observation of any praseodymium or neodymium isotopes [97]. A modified version of the nuclide chart

Table 2. Nuclides only reported in proceedings or internal reports until the end of 2011. The nuclide, author, year, laboratory, conference or report and reference of the discovery are listed.

Nuclide(s)	Author	Year	Laboratory	Conf./Report	Ref.
^{95}Cd,^{97}In[a]	R Krücken	2008	GSI	Nucl. Phys. and Astrophys.: From Stable Beams to Exotic Nuclei, 25–30 June 2008, Cappadocia (Turkey)	[102]
^{155}Pr[a] ^{156}Pr	S Czajkowski *et al*	1996	GSI	ENAM'95, 19–23 June 1995, Arles (France)	[96]
^{126}Nd	G A Souliotis	2000	MSU	Int. Conf. on Achievements and Perspectives in Nuclear Structure, 11–17 July 1999, Aghia Palaghia, Crete (Greece)	[103]
^{157,158}Nd[a]	S Czajkowski *et al*	1996	GSI	ENAM'95, 19–23 June 1995, Arles (France)	[96]
^{136}Gd,^{138}Tb	G A Souliotis	2000	MSU	Int. Conf. on Achievements and Perspectives in Nuclear Structure, 11–17 July 1999, Aghia Palaghia, Crete (Greece)	[103]
^{143}Ho	G A Souliotis	2000	MSU	Int. Conf. on Achievements and Perspectives in Nuclear Structure, 11–17 July 1999, Aghia Palaghia, Crete (Greece)	[103]
	D Seweryniak *et al*	2002	LBL	Annual Report	[104]
^{144}Tm	K P Rykaczewski *et al*	2004	ORNL	Nuclei at the Limits, 26–30 July 2004, Argonne, Illinois (USA)	[105]
	R Grzywacz *et al*			ENAM2004, 12–16 September 2004, Pine Mountain, Georgia	[106]
	C R Bingham *et al*			CAARI2004, 10–15 October 2004, Fort Worth, Texas (USA)	[107]
^{178}Tm[a]	Zs Podolyak *et al*	1999	GSI	2nd Int. Conf. Fission and Properties of Neutron-Rich Nuclei, 28 June–3 July 1999, St Andrews (Scotland)	[108]
^{150}Yb	G A Souliotis	2000	MSU	Int. Conf. on Achievements and Perspectives in Nuclear Structure, 11–17 July 1999, Aghia Palaghia, Crete (Greece)	[103]
^{181}Yb[a]	Zs Podolyak *et al*	1999	GSI	2nd Int. Conf. Fission and Properties of Neutron-Rich Nuclei, 28 June–3 July 1999, St Andrews (Scotland)	[108]
^{182}Yb[a]	S D Al-Garni *et al*	2002	GSI	Annual Report	[109]
^{153}Hf	G A Souliotis	2000	MSU	Int. Conf. on Achievements and Perspectives in Nuclear Structure, 11–17 July 1999, Aghia Palaghia, Crete (Greece)	[103]
^{164}Ir	H Kettunen *et al*	2000	Jyväskylä	XXXV Zakopane School of Physics, 5–13 September 2000, Zakopane (Poland)	[110]
	H Mahmud *et al*	2001	ANL	ENAM2001, 2–7 July 2001, Hämeenlinna (Finland)	[111]
	D Seweryniak *et al*			Frontiers of Nuclear Structure, 29 July–2 August 2002, Berkeley, California (USA)	[112]
^{234}Cm	P Cardaja *et al*	2002	GSI	Annual Report	[113]
	J Khuyagbaatar *et al*	2007	GSI	Annual Report	[114]
	D Kaji *et al*	2010	RIKEN	Annual Report	[115]
^{235}Cm	J Khuyagbaatar *et al*	2007	GSI	Annual Report	[114]
^{234}Bk	K Morita *et al*	2002	RIKEN	Frontiers of Collective Motion, 6–9 November 2002, Aizu (Japan)	[116]
	K Morimoto *et al*			Annual Report	[117]
	D Kaji *et al*	2010	RIKEN	Annual Report	[115]
^{252,253}Bk	S A Kreek *et al*	1992	LBL	Annual Report	[118]
^{262}No	R W Lougheed *et al*	1988	LBL	Annual Report	[119]
				50 years with nuclear fission, April 25–28, 1989, Gaithersburg, Maryland (USA)	[120]
	E K Hulet			Internal Report	[121]
^{261}Lr	R W Lougheed *et al*	1987	LBL	Annual Report	[122]
	E K Hulet			Internal Report	[121]
	R A Henderson *et al*	1991	LBL	Annual Report	[123]
^{262}Lr	R W Lougheed *et al*	1987	LBL	Annual Report	[122]
	E K Hulet			Internal Report	[121]
	R A Henderson *et al*	1991	LBL	Annual Report	[123]
^{255}Db	G N Flerov	1976	Dubna	3rd Int. Conf. on Nuclei Far from Stability, 19–26 May 1976, Cargese, Corsica (France)	[124]

[a] Discovered in 2012, see discussion in section 5.

showing these nuclei was included in two further publications [98, 99].

These two neodymium isotopes (^{157,158}Nd) have recently been reported (see section 5) by van Schelt *et al* [100] and Kurcewicz *et al* [101], respectively.

Another argument for not giving full credit for a discovery reported in conference proceedings are contributions from single authors (for example [102, 103]). These experiments typically involve fairly large collaborations and it is not clear that these single-author papers were fully vetted by the collaboration. Also everyone involved in the experiment and the analysis should get the appropriate credit.

The authors of the more recent proceedings and reports are encouraged to fully analyze the data and submit their final results for publication in referred journals.

4.2. Medium-mass proton rich nuclides

The proton dripline has been crossed in the medium-mass region between antimony and bismuth (Z = 51–83)

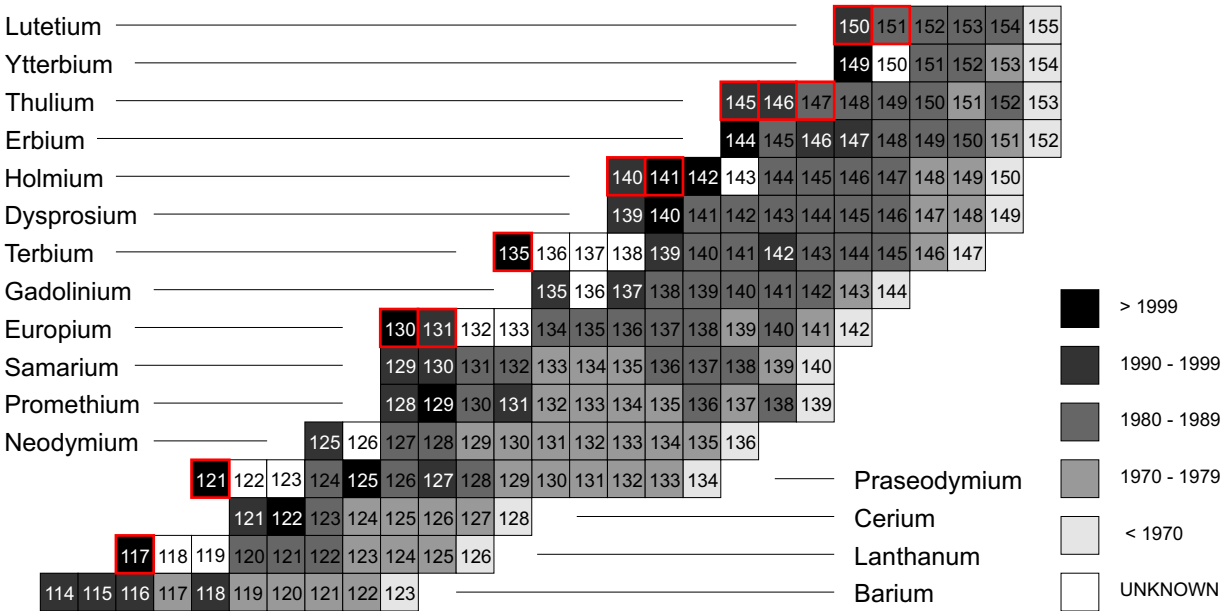

Figure 3. Chart of nuclides for neutron-deficient nuclides between barium and lutetium ($Z = 56$–71. The gray-scale coding refers to the decade of discovery. Proton emitters are identified by the thick (red) borders.

with the observation of proton emitters of odd-Z elements. Promethium is the only odd-Z element in this mass region where no proton emitters have been discovered yet. In these experiments the protons are detected in position sensitive silicon detectors correlated with the implantation of a fusion–evaporation residue after a mass separator. The high detection efficiency for these protons makes this method very efficient and nuclides far beyond the proton dripline with very small cross sections can be identified.

In contrast, for nuclides closer to the dripline proton emission is not the dominant decay mode due to the smaller Q-values for the proton decay. The identification of these nuclides is more difficult because of the lower detection efficiency for β- and γ-rays. In fact many of these nuclei were identified by β-delayed proton emission from excited states of the daughter nuclei. Thus, there are isotopes not yet discovered between the lightest β-emitters and the heaviest proton emitters for the odd-Z elements.

Figure 3 shows the medium-mass neutron-deficient region of the chart of nuclides. The thick (red) borders indicate proton emitters and the gray shades of the nuclides indicate the decade of discovery.

Currently nine odd-Z nuclides (^{118,119}La, ^{122,123}La, ^{132,133}Eu and ^{136,137,138}Tb) fall into these gaps. For the tenth missing nuclide, ^{143}Ho, ^{142}Ho has already been identified by β-delayed proton emission [125]. In fact, decay properties of ^{143}Ho have also been measured but the results were only reported in an annual report [104].

There are three even-Z holes in this mass region: ^{126}Nd, ^{136}Gd and ^{150}Yb. In all three cases, the even more neutron-deficient nuclides were observed by the detection of β-delayed proton emission at the Institute of Modern Physics, Lanzhou, China (^{125}Nd [126], ^{135}Gd [127] and ^{149}Yb [128]).

The identification of ^{126}Nd, ^{136}Gd and ^{150}Yb in the fragmentation reaction of a 30 MeV/nucleon ^{197}Au beam has been reported only in a contribution to a conference proceeding [103]. The recent advances in beam intensities and detection techniques for fragmentation reactions (especially identification and separation of charge states) should make it possible to discover these and many more additional nuclides along and beyond the proton dripline in this mass region.

4.3. Medium-mass neutron-rich nuclei

In contrast to the proton dripline the neutron dripline has not been reached for medium mass nuclides. The heaviest neutron-rich nuclide shown to be unbound is ^{39}Mg [129]. Most of the most neutron-rich nuclides have been produced in projectile fragmentation or projectile fission over the last 15 years. The nuclides are separated with fragment separators according to their magnetic rigidity (= momentum over charge of the nuclides which corresponds approximately to their A/Z) and identified by time-of-flight and energy-loss measurements.

Figure 4 displays the neutron-rich part of the chart of nuclides between argon and thorium ($Z = 18$–90) as a function of A/Z. It shows the A/Z ranges covered by the different experiments. The figure also includes the most recent measurement by Kurcewicz *et al* [101] (see section 5). If one considers that the location of the neutron dripline is predicted to be more or less constant at about 3.2 in this mass region, it is clear from the figure that it is still far away. The limits of the projectile fragmentation/fission method are currently determined by the small cross sections which can be overcome to an extend by improvements of primary beam intensities and/or larger acceptance separators. In the long term the method is limited by the limited availability of neutron-rich projectiles.

4.4. Superheavy nuclides

The discovery of superheavy nuclides has always been special because it is directly related to the discovery of new elements.

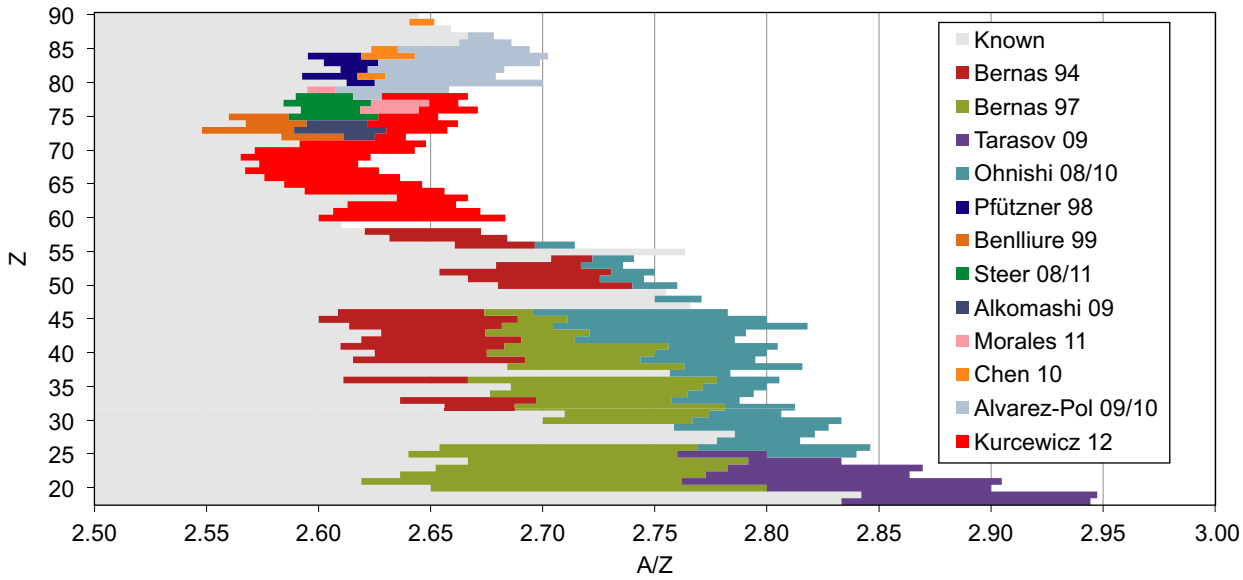

Figure 4. Neutron-rich nuclei between argon and thorium discovered by projectile fragmentation or projectile fission as a function of A/Z. The data as labeled in the figure are from Bernas 94 [97], Bernas 97 [98], Tarasov 09 [130], Ohnishi 08/10 [57, 131], Pfützner 98 [132], Benlliure 99 [133], Steer 08/11 [134, 135], Alkomashi 09 [136], Morales 11 [137], Chen 10 [53], Alvarez-Pol 09/10 [54, 138] and Kurcewicz [101]. The legend in the figure refers to the first author and year of the publications.

It is interesting to follow the evolution of element discovery and the discovery of nuclides. In the 1990 book 'The elements beyond uranium' Seaborg and Loveland showed the number of discovered transuranium elements and nuclides as a function of year [139]. Figure 5 displays an extension of these data until today. The number of discovered nuclides tracks closely the number of discovered elements with about 10 isotopes per elements.

In addition to the efforts to discover elements 119 and 120 it is important to link the isotopes of the elements beyond 113 to known nuclides. Figure 6 shows the nuclear chart beyond nobelium. It shows the separation of the more neutron-rich nuclides up to $Z = 118$ produced in 'hot' fusion–evaporation reactions from the less neutron-rich nuclides up to $Z = 113$ which were predominantly produced in 'cold' fusion–evaporation reactions. No isotone with $N = 164$ has been observed so far which does not mean that this isotone line corresponds to the separation of the decay chains.

Table 3 lists the ten currently observed unconnected decay chains. There are five even-Z and five odd-Z chains. The four chains starting at ^{282}113, ^{285}Fl, ^{288}115 and ^{291}Fl bridge the $N = 164$ gap and end in ^{270}Bh, ^{265}Rf, ^{268}Db and ^{267}Rf, respectively. It should be mentioned that the odd-Z $N-Z = 57$ chain passes through the $N = 164$ isotone ^{271}Bh; however, the properties of this nuclide could not unambiguously be determined [95].

The decay chains cannot be connected to known nuclides by extending them to lower masses because they terminate in nuclides which spontaneously fission. The relationship has to be established by systematic features of neighboring isotopes for different elements. Thus the missing isotopes $^{279-281}$113, $^{278-280}$Cn, $^{275-277}$Rg and $^{274-276}$Ds as well as the other N = 164 isotopes ^{273}Mt, ^{272}Hs, ^{271}Bh, ^{270}Sg and ^{269}Db have to measured. In total there are 39 nuclides still to be discovered between already known light and heavy rutherfordium and $Z = 113$ nuclides.

In addition, there are a few gaps of unknown nuclides in the lighter (trans)uranium region. ^{239}Bk and the two curium isotopes ^{234,235}Cm have yet to be discovered, although as mentioned in section 4.1 the curium isotopes have been reported in annual reports. Also three uranium isotopes are still unknown, ^{220,221}U and ^{241}U. It is especially surprising that the two lighter isotopes ^{220,221}U have not been observed because three even lighter isotopes ($^{217-219}$U) are known. ^{222}U was formed in the fusion–evaporation reaction ^{186}W(^{40}Ar,4n) [140] and most of the other light uranium isotopes were formed in 4n or 5n reactions. Thus ^{220,221}U should be able to be populated and identified in ^{184}W(^{40}Ar,4n) and ^{186}W(^{40}Ar,5n) reactions, respectively.

4.5. Beyond the driplines

As mentioned in section 3 the present definition of nuclides also includes very short-lived nuclides beyond the proton and neutron driplines. So far, these nuclides are only accessible in the light-mass region and characteristics of many of these nuclides up to magnesium beyond the proton dripline and up to oxygen beyond the neutron dripline have been measured.

The proton dripline has most likely been reached or crossed for all elements up to technetium ($Z = 43$). Table 4 lists the first isotope of elements between aluminum and technetium which has been shown to be unbound but which has not been identified yet or the first isotope for which nothing is known, so that in principle it still could be bound or could have a finite lifetime. With maybe the exception of scandium, bromine and rubidium where resonances have been already measured for ^{38}Sc [141], ^{69}Br [142] and ^{73}Rb [143], resonance parameters for at least one isotope of these elements should be in reach in the near future.

For elements lighter than aluminum at least one unbound isotope has been identified. Although not impossible it is

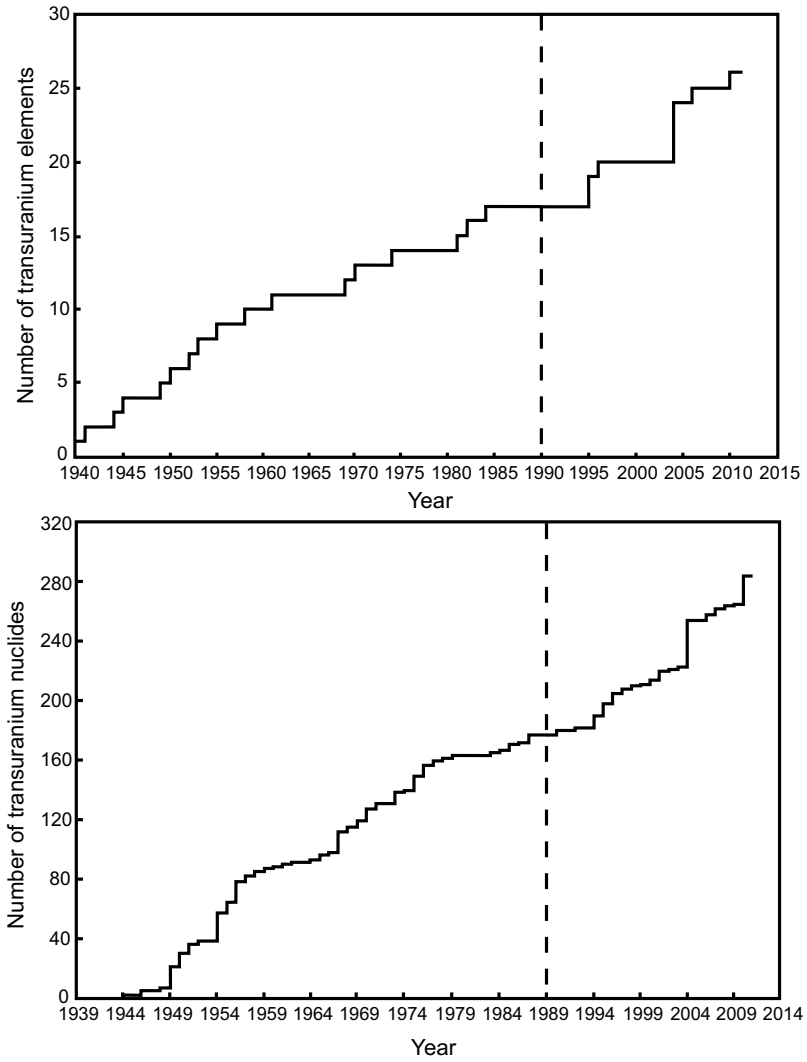

Figure 5. Number of discovered transuranium elements and nuclides. The data until 1989/1990 indicated by the dashed line were taken from [139].

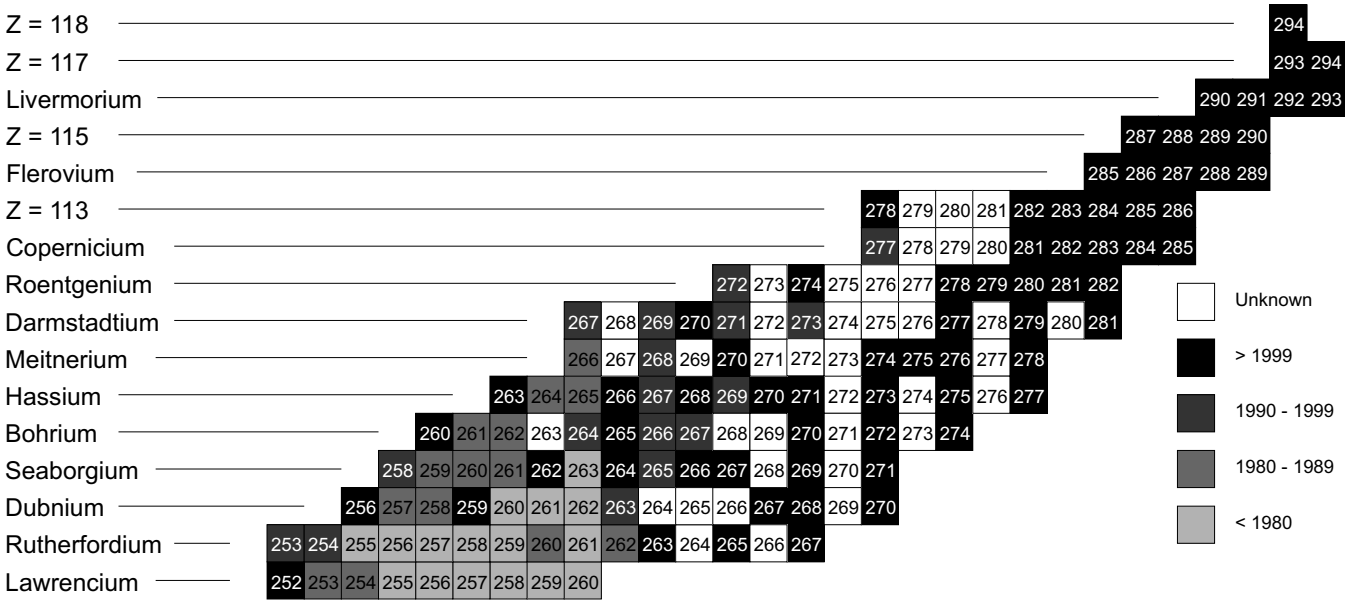

Figure 6. Chart of nuclides for elements heavier than nobelium. The gray-scale coding refers to the decade of discovery.

Table 3. Unconnected superheavy decay chains. The (N–Z) value, first and last nuclides and the number of α-decays in the chains are listed.

	(N–Z) Chain	First	Last	of α decays
Even-Z	57	^{285}Fl	^{265}Rf	5
	58	^{294}118	^{282}Cn	3
	59	^{291}Lv	^{267}Rf	6
	60	^{292}Lv	^{284}Cn	2
	61	^{293}Lv	^{277}Hs	4
Odd-Z	56	^{282}113	^{266}Db	4
	57	^{287}115	^{267}Db	5
	58	^{288}115	^{268}Db	5
	59	^{293}117	^{281}Rg	3
	60	^{294}117	^{270}Db	6

unlikely that further nuclides will exist for which characteristic resonance parameters can be measured.

For neutron-rich nuclei characteristic properties of at least two isotopes beyond the neutron dripline have been identified for the lightest elements, hydrogen, helium and lithium. Neutron-rich nuclides between beryllium and magnesium which have been shown or expected to be unbound but have not been observed are listed in table 5. Most of these nuclides should be able to be measured in the near future. In fact, ^{16}Be, ^{26}O and ^{28}F have been discovered recently (see section 5). The open question whether the ($A - 3Z = 6$) nuclei between fluorine and magnesium (^{33}F, ^{36}Ne, ^{39}Na and ^{42}Mg) should be answered in the near future with the available increased intensities of the RIBF at RIKEN [56]. Beyond aluminum the dripline has most likely not been reached yet with the observation that ^{42}Al is bound with respect to neutron emission [151].

5. New discoveries in 2012

While in 2010 a record number of 110 new nuclei were reported [159], only seven additional new nuclei were discovered in 2011. The trend was again reversed in 2012 with the new identification of up to 67 nuclei. Kurcewicz *et al* alone reported 59 new neutron-rich nuclei between neodymium and platinum [101]. These include ^{158}Nd, ^{178}Tm and ^{181,812}Yb which had previously been reported only in conference proceedings (see section 4.1). Kurcewicz *et al* reported the discovery of 60 new nuclides; however, ^{157}Nd was reported in a paper by Van Schelt *et al* [100] which had been submitted five months earlier. Van Schelt also measured ^{155}Pr for the first time; both isotopes had previously been reported in a conference proceeding. In addition, resonances in the light neutron-unbound nuclei ^{16}Be [160], ^{26}O [161] and ^{28}F [162] were measured for the first time.

The remaining three nuclides, ^{95}Cd, ^{97}In and ^{99}Sn, bring up the discussion of what should be counted as a discovery. The particle identification plot in the recent publication by Hinke *et al* exhibits clear evidence for the presence of ^{95}Cd and ^{97}In and a few events of ^{99}Sn [163]. However, neither the text nor the figure caption mentions the discovery of these nuclides. In an earlier contribution to a conference proceeding Krücken reported the discovery of ^{95}Cd and ^{97}In, but not ^{99}Sn, from the same experiment [102].

In addition to these 66 nuclides another six new nuclides (^{64}Ti, ^{67}V, ^{69}Cr, ^{72}Mn, ^{70}Cr and ^{75}Fe) were reported in a contribution to a conference proceeding [164].

6. Long term future

Over 3000 different isotopes of 118 elements are currently known. In a recent paper theoretical calculations revealed that about a total of 7000 bound nuclei should exist, thus more than double the nuclides currently known [1]. However, not all will ever been in reach as can be seen in figure 7. The figure shows the known nuclides first produced by light–particle reactions, fusion/evaporation reactions, and spallation/fragmentation which are shown in green, orange and dark blue, respectively. Nuclides of the radioactive decay chains are shown in purple and stable nuclides in black. The yellow regions show unknown nuclides predicted by [1]. The light-blue border corresponds to the uncertainty of the driplines in the calculations.

In the region of $Z > 82$ and $N > 184$ alone about 2000 nuclides will most probably never be created. If one conservatively adds another 500 along the neutron dripline in the region above $Z \sim 50$ it can be estimated that another approximately 1500 nuclides (7000 predicted minus 3000 currently known minus 2500 out of reach) are still waiting to be discovered. In the 2004 review paper on the limits of nuclear stability it was estimated that the rare isotope accelerator (RIA) which had been proposed at the time would be able to produce about 100 new nuclides along the proton dripline below $Z \sim 82$ [67]. Since then only about 20 of these nuclides have been observed. Thus the next generation radioactive beam facilities (the Radioactive Ion-Beam Factory RIBF at RIKEN [165], the Facility for Antiproton and Ion Research FAIR at GSI [166], and the Facility for Rare Isotope Beams FRIB at MSU [167, 168]) should be able to produce approximately 80 new neutron-deficient nuclides. Equally critical for new discoveries at these facilities are the next generation fragment separators, BIG-RIPS [57, 169], the Super FRS [170], and the FRIB fragment separator [171], respectively.

Along the neutron dripline RIA was estimated to make another 400 nuclides below $Z \sim 50$ [67] of which about 70 have been discovered in the meantime leaving about another 330 for the new facilities in the future.

The remaining unkown nuclides in the various regions of the nuclear chart have to be produced by different reaction mechanisms. Projectile fragmentation reactions will most likely be utilized to populate neutron-deficient nuclides below $Z \sim 50$ and for nuclides above $Z \sim 82$ fusion–evaporation reactions are the only possibility. The use of fusion–evaporation reactions with radioactive beams might be an alternative to reach nuclides which cannot be populated with stable target-beam combinations [67]. Neutron-deficient nuclides in the intermediate mass region ($50 < Z < 82$) have been produced so far by fusion–evaporation reactions; however, projectile fragmentation could be a viable alternative [103].

New neutron-rich nuclides below $Z \sim 82$ will most likely be only reachable by projectile fragmentation/fission reactions.

Table 4. Nuclides beyond the proton dripline which have been demonstrated to be unbound or have not been reported yet.

Z	Nuclide	Author	Year	Laboratory	Comments	Ref.
13	^{21}Al				Not measured	
14	^{21}Si				Not measured	
15	^{25}P	M Langevin *et al*	1986	GANIL		[144]
16	^{26}S	A S Fomichev *et al*	2011	Dubna		[145]
17	^{29}Cl	M Langevin *et al*	1986	GANIL		[144]
	^{30}Cl	M Langevin *et al*	1986	GANIL		[144]
18	^{30}Ar				Not measured	
19	^{33}K	M Langevin *et al*	1986	GANIL		[144]
	^{34}K	M Langevin *et al*	1986	GANIL		[144]
20	^{34}Ca				Not measured	
21	^{38}Sc				Not measured, but ^{39}Sc unbound	
22	^{38}Ti	B Blank *et al*	1996	GANIL		[146]
23	^{42}V	V Borrel *et al*	1992	GANIL		[147]
24	^{41}Cr				Not measured	
25	^{44}Mn	V Borrel *et al*	1992	GANIL		[147]
	^{45}Mn	V Borrel *et al*	1992	GANIL		[147]
26	^{44}Fe				Not measured, but ^{45}Fe 2p emitter	
27	^{49}Co	B Blank *et al*	1994	GANIL		[148]
28	^{47}Ni				Not measured, but ^{48}Ni 2p emitter	
29	^{54}Cu	B Blank *et al*	1994	GANIL		[148]
30	^{53}Zn				Not measured, but ^{54}Zn 2p emitter	
31	^{59}Ga	A Stolz *et al*	2005	MSU		[149]
32	^{59}Ge				Not measured	
33	^{63}As	A Stolz *et al*	2005	MSU		[149]
34	^{63}Se				Not measured	
35	^{68}Br				Not measured, but ^{69}Br unbound	
36	^{68}Kr				Not measured	
37	^{72}Rb				Not measured, but ^{73}Rb unbound	
38	^{72}Sr				Not measured	
39	^{75}Y				Not measured	
40	^{77}Zr				Not measured	
41	^{81}Nb	Z Janas *et al*	1999	GANIL		[150]
42	^{82}Mo				Not measured	
43	^{85}Tc	Z Janas *et al*	1999	GANIL		[150]

Table 5. Nuclides beyond the neutron dripline which have been demonstrated to be unbound or have not been reported yet.

Z	Nuclide	Author	Year	Laboratory	Comments	Ref.
4	^{15}Be	A Spyrou *et al*	2011	MSU		[152]
	^{16}Be[a]	T Baumann *et al*	2003	MSU		[153]
5	^{20}B	A Ozawa *et al*	2003	RIKEN		[154]
	^{21}B	A Ozawa *et al*	2003	RIKEN		[154]
6	^{21}C	M Langevin *et al*	1985	GANIL		[155]
	^{23}C				Not measured, but ^{21}C unbound	
7	^{24}N	H Sakurai *et al*	1999	RIKEN		[156]
	^{25}N	H Sakurai *et al*	1999	RIKEN		[156]
8	^{26}O[a]	D Guillemaud-Mueller *et al*	1990	GANIL		[157]
	^{27}O	O Tarasov *et al*	1997	GANIL		[158]
	^{28}O	O Tarasov *et al*	1997	GANIL		[158]
9	^{28}F[a]	H Sakurai *et al*	1999	RIKEN		[156]
	^{30}F	H Sakurai *et al*	1999	RIKEN		[156]
	^{32}F				Not measured, but ^{30}F unbound	
	^{33}F				potentially bound	
10	^{33}Ne	M Notani *et al*	2002	RIKEN		[129]
	^{35}Ne				Not measured, but ^{33}Ne unbound	
	^{36}Ne				potentially bound	
11	^{36}Na	M Notani *et al*	2002	RIKEN		[129]
	^{38}Na				Not measured, but ^{36}Na unbound	
	^{39}Na				potentially bound	
12	^{39}Mg	M Notani *et al*	2002	RIKEN		[129]
	^{41}Mg				Not measured, but ^{39}Mg unbound	
	^{42}Mg				potentially bound	

[a] Discovered in 2012, see section 5.

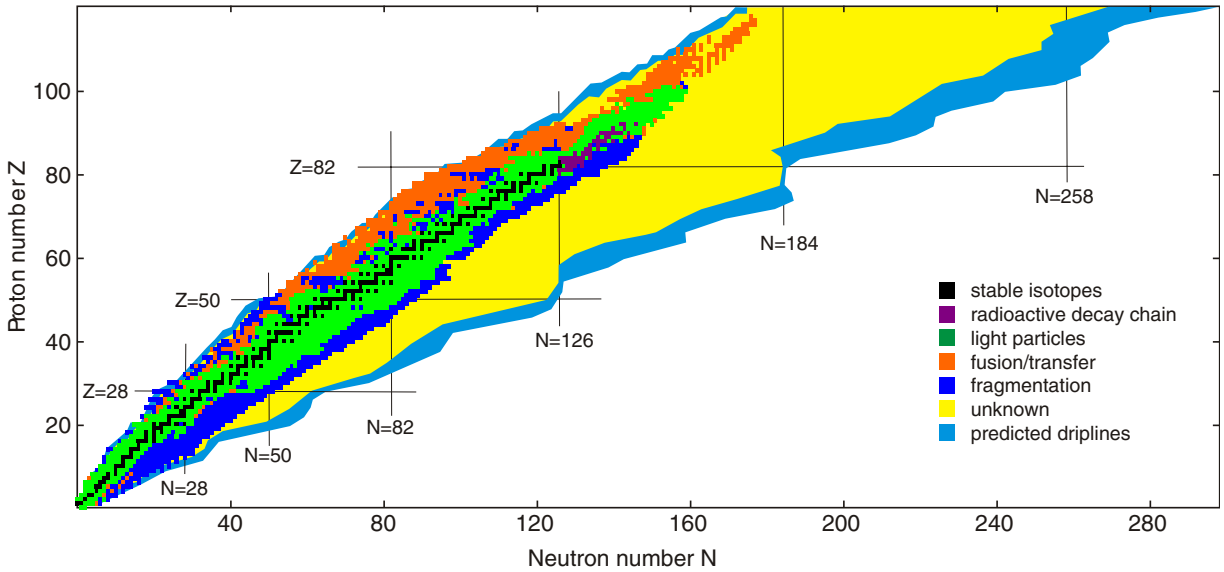

Figure 7. Chart of nuclides. Stable nuclides are shown in black. The other known nuclides are grouped according to the production mechanism of their discovery: radioactive decay chains (purple), light-particle induced reactions (green), fusion/transfer reactions (orange), and spallation or projectile fragmentation/fission (dark blue). Nuclides predicted to exist according to [1] are shown in yellow where the light-blue area shows the uncertainty of the driplines.

The 2004 review predicted that the dripline would be reachable up to $Z \sim 30$ [67]. If the dripline is as far away as estimated in the recent calculations [1] it could be that the dripline will not be reached beyond $Z \sim 16$; at least not in the near future.

The search for new superheavy elements and therefore also new nuclei continues to rely on fusion–evaporation reactions [172–174]. However, recent calculations suggest that deep inelastic reactions or multi-nucleon transfer reactions on heavy radioactive targets (for example ^{248}Cm) might be a good choice to populate heavy neutron-rich nuclei [175–177]. The use of radioactive beams on radioactive targets could also be utilized for fusion–evaporation reactions in the future [177, 178].

7. Conclusion

The quest for the discovery of nuclides that never have been made on Earth continues to be a strong motivation to advance nuclear science toward the understanding of nuclear forces and interactions. The discovery of a nuclide is the first necessary step to explore its properties. New discoveries have been closely linked to new technical developments of accelerators and detectors. In the future it will be critical to develop new techniques and methods in order to further expand the chart of nuclides.

The discovery potential is not yet limited by the number of undiscovered nuclides. About 1500 could still be created. This would correspond to about 90% of all predicted nuclides below $N \sim 184$ which should be sufficient to constrain theoretical models to reliably predict properties of all nuclides as well as the limit of existence.

Acknowledgments

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