THE LEGACY OF MARIA SKŁODOWSKA-CURIE HUNDRED YEARS AFTER THE DISCOVERY OF THE ATOMIC NUCLEUS*

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With the discovery of the first radioactive chemical elements Maria Skłodowska-Curie made the step into the *terra incognita* of radioactive elements and isotopes. With the first quantitative measurement of nuclear radiation and the development of radiochemical separation she discovered polonium and radium. The alpha radiation was used for discoveries forming our picture of the atomic nucleus. She found that radiation originating from nuclear decay allows a look into the atomic nucleus. In this contribution, we will follow the discoveries, initiated by the work of Maria Skłodowska-Curie, on the way to present nuclear physics with modern rare-isotope facilities. Her legacy includes medical application of radiation, now being an integral part of modern nuclear medicine.

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1. The birth of nuclear physics

With the discovery of X-rays emitted from cathode rays Wilhelm Conrad Röntgen discovered in 1895 "a new kind of rays" [1]. One year later, Antoine Henry Becquerel discovered that a radiation is emitted from uranium salts [2]. He called it "a new property of matter". These two discoveries mark the first steps into a new field of physics. Maria Curie started the systematic investigation of this new phenomenon and asked the question, "what is the origin of this radiation?" Her step from radiation detection to precise measurement and investigation of this radiation started the field of

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nuclear physics, the investigation of the atomic nucleus as the building block of elementary matter. The first milestone was the discovery of the first unstable chemical elements in 1898: polonium *On a New Radioactive Substance Contained in Pechblende* [3] and radium: *On a New Strong Radioactive Substance Contained in Pechblende* [4]. A historical overview of nuclear physics as a table, listed with year, discoverer, discovery, and publication is given in [5] and was used by the author.

The key to the discoveries was the development of radiochemical methods: the continuous enrichment of the radioactive substance in subsequent steps of chemical separation. This was only possible with a precise measurement of radioactivity with sophisticated instrumentation: an ionization chamber where on one of the plates the radioactive substance was deposited, a highly sensitive quadrant electrometer and the piezo balance (Fig. 1). The ionizing radiation from the radioactive substance placed on one of the condenser plates discharged it. The discharge was measured with the piezo balance using the quadrant electrometer as indicator for the compensation voltage.

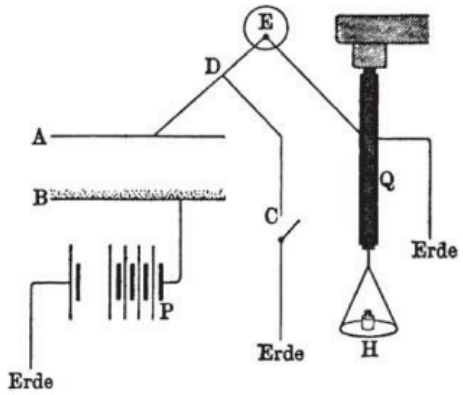


Fig. 1. The first instrument used in nuclear physics: The setup used by Maria Curie in her thesis to measure radioactivity. A, B: Condenser plates forming the ionization chamber, B loaded with radioactive substance, E: electrometer, Q: piezo balance with weight H [6].

In 1910 Maria Curie and Andre Debierne separated 2 mg of a substance containing 5% of ^{210}Po from the remains of about two tons of pechblende. The separation of radium was a challenge: From several tons of pechblende they separated about 100 mg of radium by continuous chemical enrichment of the radioactive fraction [7]. Finally, polonium was separated by fractional crystallization, a method later used also by Strassmann in the discovery of nuclear fission. With her work Maria Skłodowska-Curie laid the basis for:

— *Nuclear chemistry and physics.*
Creation and an investigation of new elements and isotopes.
Structure research.

— *Nuclear reactions.*
Nuclear shape.
Transmutation of chemical elements.

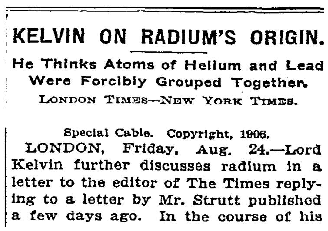
— *Application.*
Nuclear medicine.

She paved the way to modern nuclear physics [8] and nuclear medicine.

1.1. New phenomena, questions, and concerns

The discovery showed an unusual behavior of matter raising many questions: "In this manner a source of light is obtained which is very feeble, to tell the truth, but which operates without a source of energy. Here is at least an apparent contradiction to Carnot's Principle" [4]. What is the original source of the energy of the radiation?

An even more fundamental question was the transmutation in radioactive decays. "Chemical elements, the building blocks of matter, can be destructed or produced by a distinct type of radioactivity". In 1661 Robert Boyle had endorsed the view of chemical elements as the undecomposable constituents of material bodies. This had been generally accepted and the recent consequence was Mendelejevs periodic table in 1869. So it was only natural that there was opposition against this new way of thinking, most prominent from Lord Kelvin in the "New York Times", 1906, Fig. 2.



KELVIN ON RADIUM'S ORIGIN.

He Thinks Atoms of Helium and Lead Were Forcibly Grouped Together.

LONDON TIMES—NEW YORK TIMES.

Special Cable. Copyright, 1906.

LONDON, Friday, Aug. 24.—Lord Kelvin further discusses radium in a letter to the editor of The Times replying to a letter by Mr. Strutt published a few days ago. In the course of his

Fig. 2. Lord Kelvin opposing the idea of transmutation in nuclear decay.

There were also thoughts about nuclear energy and dangers from this discovery. In his Nobel Lecture in 1903 Pierre Curie says:

"The quantity of heat released by radium in several years is enormous if it is compared with the heat released in any chemical reaction with the same weight of matter".

"Radium rays have been used in the treatment of certain diseases. In certain cases their action may become dangerous ..."

"It can be even thought that radium could become very dangerous in criminal hands, and there the question can be raised whether mankind benefits from knowing the secrets of nature or whether this knowledge will be harmful for it."

The discoveries of new elements continued. In 1918, Lise Meitner and Otto Hahn, and independently Frederick Soddy and John Cranston, discover "protoactinium", the precursor of actinium, the last unknown of the radioactive elements descending from uranium. Later IUPAC changed the name to "protactinium".

1.2. Radium and thorium rays the source for discoveries, forming our picture of elementary matter

Alpha rays from polonium and radium played a crucial role in the discoveries forming our picture of the atomic nucleus. Ernest Rutherford in 1909 discovered the atomic nucleus by alpha scattering on a thin gold foil [9]. In 1919 he made two important discoveries in one experiment [10]. In the first nuclear reaction

$$^{14}\mathrm{N} + \alpha \rightarrow {}^{17}\mathrm{O} + \text{proton}$$

he observed for the first time the transmutation of a chemical element and discovered the proton as a constituent of the atomic nucleus. It lasted until 1932 that James Chadwick discovered the neutron. He generated the neutrons by α particles of polonium hitting beryllium target in vacuum to avoid scattering of the α particles. His paper [11] starts: "It has been shown by Bothe and others that beryllium when bombarded by α-particles of polonium emits a radiation of great penetrating power, which has an absorption coefficient in lead of about 0.3 $(\mathrm{cm})^{-1}$. Recently Mme. Curie-Joliot and M. Joliot found, when measuring the ionization produced by this beryllium radiation in a vessel with a thin window, that the ionization increased when matter containing hydrogen was placed in front of the window ...". Six years later, Otto Hahn and Fritz Strassmann use Chadwicks neutron generator to irradiate ^{238}Uranium to produce transuranium elements using Enrico Fermi's method of neutron irradiation and subsequent β^- decay converting a neutron into a proton to climb up the chart of nuclides by one Z unit

$$^{238}\mathrm{U} + n \rightarrow {}^{239}\mathrm{U} \rightarrow {}^{239}93 + \beta^- \,.$$

Instead they found lighter elements, barium and masurium, now called technetium. In fact, the result was not clear from the beginning. As the chemistry of polonium and barium is quite similar, they first believed in finding new polonium isotopes, formed by α-chains starting from the uranium

after neutron capture [12]. Immediately after a notice from Otto Hahn about his discovery, Lise Meitner and Otto Frisch explained this result as nuclear fission [13]. They calculated the tremendous energy release in the fission process and predicted the termination of the table of elements near $Z = 100$ by fission, limited by the large nuclear charge creating great Coulomb forces, increasing with Z^2 and finally destroying the heavy nucleus. Not much later the first transuranium element, neptunium, was discovered by Philip H. Abelson and Edwin M. McMillan [14]. In the following time, the elements plutonium ($Z = 94$), americium ($Z = 95$), and curium were discovered, created by successive neutron capture and β^- decay [15, 16].

2. A new era starts: accelerators in nuclear physics

When John Cockcroft and Ernest Walton started the operation of the first particle accelerator in 1932, a new era of nuclear physics started. With protons accelerated to 700 keV, they observed the spallation of lithium isotopes [17]. Already in the discovery of berkelium by Thompson *et al.* in 1950 heavy ion acceleration played a role. Berkelium was produced in the heavy-ion fusion reaction

$$^{241}\mathrm{Am} + {}^{4}\mathrm{He} \rightarrow {}^{243}\mathrm{Bk} + 2n\,.$$

This type of fusion reaction, light projectiles, typically isotopes of oxygen or nitrogen and actinide targets bred in the nuclear reactor, led to the discoveries of new elements including seaborgium ($Z = 106$).

2.1. Towards superheavy elements

With the addition of microscopic effects to the nuclear droplet, the question arose as to whether "superheavy" nuclei, can exist at the next doubly magic shell closure after ^{208}Pb beyond the droplet limit estimated by Meitner and Frisch only due to shell stabilization [19]. Most of the early predictions place the island around $Z = 114$ and $N = 184$. Modern calculations with self-consistent models place the magic proton number at $Z = 120$ or even $Z = 126$ [20]. At the same time the progress in accelerator development allowed the acceleration of heavier projectiles. Taking advantage of this new development Oganessian proposed the use of the doubly magic ^{208}Pb as a target and irradiated it with appropriate projectile beams such as the most neutron rich isotopes of titanium or chromium to create cold compound systems in order to enhance the survival probability of these highly fissile elements at the top of the chart of nuclides [21]. There were however two obstacles: Superheavy nuclei were predicted as an *island* in the "sea of instability", beginning somewhere beyond rutherfordium, $Z = 104$, and the *extra*

push which forbids the fusion of massive systems at the Coulomb barrier. Both problems were solved in SHIP experiments [22]. With the discovery of hassium, $Z = 108$, a new shell region of deformed nuclei was found. It is centered at $Z = 108$ and $N = 162$ and forms the bridge from the transuranium nuclei to the predicted SHE region. The old and the new situation are schematically depicted in Fig. 3.

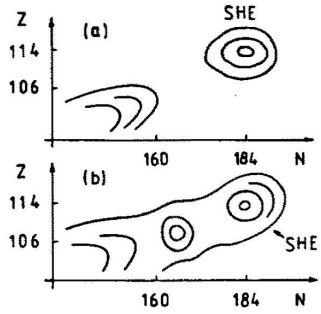


Fig. 3. The shell region around hassium, a bridge to SHE. (a) Situation before and (b) after the discovery of the shell region around ^{270}Hs. Courtesy A. Sobiczewski.

At present, two ways to produce elements beyond seaborgium ($Z = 106$) are known, Fig. 4. The cold heavy ion fusion with ^{208}Pb and ^{209}Bi targets which was successful in the discoveries of elements 107 to 112 at SHIP [22], and element 113 at RIKEN, Japan, and the hot fusion with ^{48}Ca beams and appropriate actinide targets bred in the reactor, which was successful in producing the new elements 114 to 118 at JINR Dubna [23]. Both methods were proposed and pioneered by Yu.Ts. Oganessian.

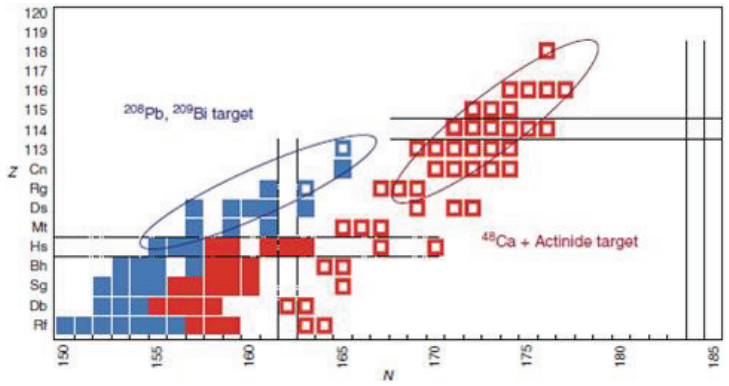


Fig. 4. Production of superheavy elements by cold (grey/blue color) and hot (dark grey/red color) fusion. The sub-shell at $Z = 108$ and $N = 162$ and the spherical shell closure 1t $Z = 114$ and $N = 184$ are indicated.

What did we learn? The idea of superheavy nuclei has been confirmed, nuclei can exist only by shell stabilization in "the sea of instability" beyond the liquid drop limit. The heaviest element reported up to now is element 118, searches for element 120 are under way at SHIP and TASCA, GSI. Though the magic number 114 has already been passed, there is no indication for spherical superheavy nuclei.

The explored region is too far away from the magic neutron number 182 (Fig. 4). Future work will concentrate on SHE chemistry to place the chemical elements in the periodic table, structure research, and, most important, the search for the spherical superheavy nuclei. Intensity upgrades of accelerators and new separators with the capability of mass identification are under way, *e.g.* the design study in the GSI–Giessen–Manipal Collaboration.

3. A new challenge: Radioactive beam facilities

The first steps into the region far-off stability were made at CERN-ISOLDE. In 1951 O. Kofoed Hansen and K.-O. Nielsen showed that it is possible to separate short-lived isotopes of noble gas elements that were produced by connecting a target, irradiated by protons, directly to an isotope separator on-line. In 1967, the 600 MeV CERN Synchrocyclotron SC started providing beams for ISOLDE. The ISOLDE program ranged from nuclear physics of unstable species to astrophysics and medical research. A strength of the ISOLDE program was the application of laser methods to nuclear physics. A most exciting example is the jump in the size in odd and even mercury atoms observed by isotope shift, an indication for a jump in shape, showing, that not all nuclei are spherical but nuclear structure may change shapes [24]. In an experiment at SHIP, Andrei Andreyev *et al.* could even observe a triplet of differently shaped spin-zero states in the atomic nucleus ^{186}Pb which can exist in: spherical, oblate, and prolate shape [25].

In-flight separation pioneered at SHIP gave access to short lived species at the limits of nuclear existence. Sigurd Hofmann discovered the first ground state proton emitter, ^{151}Lu in 1982 [26]. Ground state proton decay is a new kind of radioactivity. Two proton decay from the ground state was discovered twenty years later at the FRS at GSI by Pfützner *et al.* [27].

A new era, physics with beams of unstable nuclei of high energy, started with an experiment by Isao Tanihata *et al.* at the BEVALAC, LBL Berkeley. They produced beams of unstable light nuclei at relativistic energies by projectile fragmentation, measured absorption radii, and found that these were unusually large for ^{11}Li, a nucleus near the neutron dripline [28], which was interpreted by P.G. Hansen and Bjorn Jonson as a neutron halo: pure, dilute neutron matter surrounding the nucleus as a "neutron stratosphere" (Hansen) [29]. This exciting result, obtained with the new method of "re-

versed kinematics", where the target is stable and the nucleus of interest is the projectile, allows for the first time reaction studies with short-lived and even unbound nuclear systems. This was the birth of the rare-isotope facilities *e.g.*, GANIL, France, RARF at Riken, Japan, GSI, Germany, and NSCL at MSU, United States.

As a typical example of a first-generation RI facility the UNILAC-SIS-FRS-ESR setup at GSI Darmstadt is displayed in Fig. 5. Heavy ion beams of all chemical elements from hydrogen to uranium are accelerated by the UNILAC and injected into the Heavy-Ion Synchrotron (SIS), where they are accelerated to energies of typically 1 AGeV. They are directed onto a production target, where the radioactive nuclides are produced by projectile-fragmentation or -fission. They are separated and event-wise identified in-flight and either implanted into a detector system for nuclear spectroscopy, or injected into the Experimental Storage Ring (ESR), where they are stored and cooled, or directed to the setup for reaction studies in complete kinematics. New challenges with relativistic RI beams are:

— Single-atom in-flight identification in mass A and nuclear charge Z.

— Reactions with short lived species in complete kinematics.

— Physics with stored and electron-cooled beams and atomic nuclei.

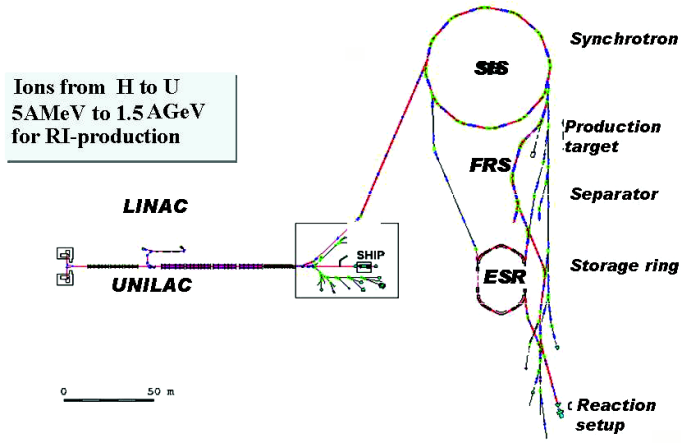


Fig. 5. The SIS-FRS-ESR facility at GSI Darmstadt, with the low-energy area with SHIP.

Reaction studies with light nuclei from helium to carbon at and beyond the drip-lines are a laboratory to study basic nuclear properties including interactions and the three body force. Halo nuclei with the neutrons moving loosely bound around the nuclear core give access to nucleons in well

defined states. In heavier nuclei with large neutron excess, a neutron skin is observed. A convincing example is the measurement of the neutron skin in the sodium isotopes by Suzuki *et al.* [30] displayed in Fig. 6. It shows the total absorption radii giving the nuclear radius and the charge radii from laser-measured isotope shifts. The difference is the neutron skin. The neutron skin is pure neutron matter. It gives access to the nuclear equation of state and the properties of the crust of neutron stars as well.

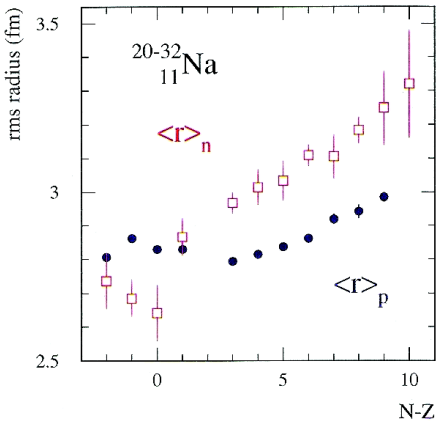


Fig. 6. Neutron skin in the neutron rich sodium isotopes, squares (red): nuclear radii measured from absorption radii, dots: charge radii from laser spectroscopy.

3.1. New shell nuclei and fission in-flight, a rich source for neutron rich species

A key issue of nuclear structure research is the nature of closed shells and doubly magic nuclei at the limits. The key question is the persistence of nuclear shells in unbalanced nuclear matter, in nuclei with large neutron- or proton-excess. Specifically, shell structure in neutron rich species along the r-process path is crucial in the creation of chemical elements in the cosmos. Major discoveries at the FRS were the doubly magic nuclei ^{100}Sn by Schneider *et al.* in 1994 [31] and of ^{78}Ni in 1995 by Engelmann *et al.* [32].

A breakthrough in the production of neutron rich species was the experiment on the fission of uranium in-flight. A beam of ^{238}U with an energy of 750 AMeV was directed onto a lead target. The fission fragments were separated and identified in-flight in A and Z event-by-event. In this experiment for the first time it was possible to observe the heavy and the light group of fission fragments in the same spectrum (Fig. 7). Uranium fission in-flight is a rich source of neutron rich nuclides. Already in this first experiment more than 40 new isotopes were identified.

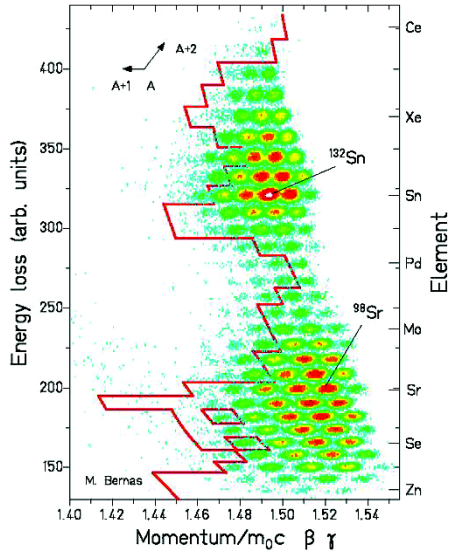


Fig. 7. The heavy and the light group of fission fragments in the in-flight fission of ^{238}U at 750 AMeV. Each dot in this plot represents an individual atomic nucleus. Please note: the most neutron rich nuclei are displayed to the right, the borderline of known species is drawn in black (red) color.

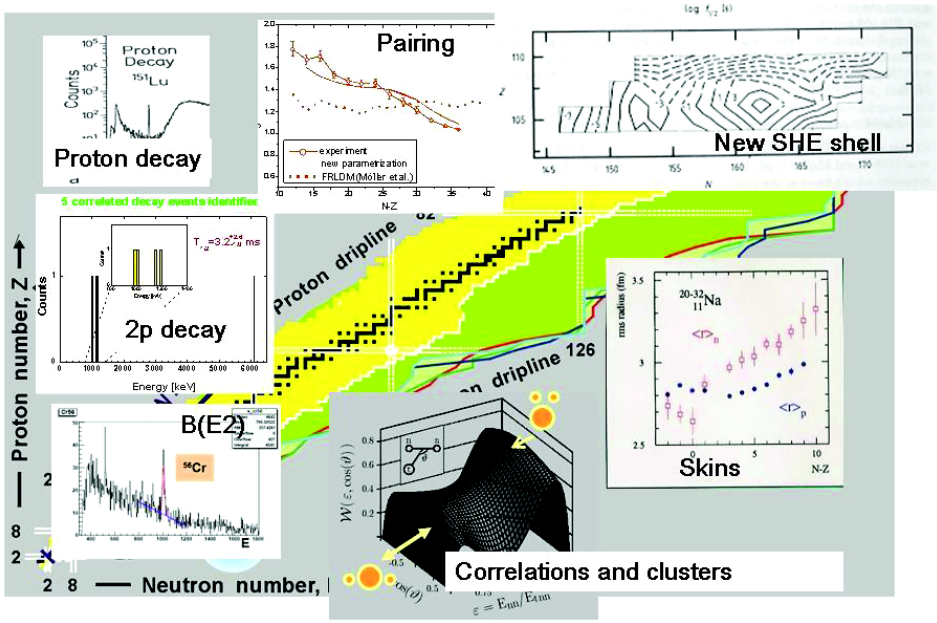


Fig. 8. Highlights of recent nuclear structure research.

3.2. Masses and half-lives of atomic nuclei

In the 1990s, a paradigm change in mass spectrometry occurred: from large mass spectrometers to trapping and cooling, or in ion optical terms: from (p, x) phase-space to (E, t) phase-space, from the measurement of line spectra to frequency measurement. This became possible through a new technique: ion cooling. The new technique has a precision of 10^{-6} or better and is sensitive to single atoms. It was pioneered at CERN ISOLDE with ISOLTRAP, an ion trap with gas cooling connected to the ISOLDE separator, and at GSI with the ESR, a storage cooler ring for ions at relativistic energies, coupled to the projectile fragment separator FRS [34]. Specifically, the ESR is well suited for large-scale inspections of the nuclear mass surface. Already in the first experiment with stored and cooled fragments of ^{209}Bi more than 500 masses were measured and more that 200 new masses could be measured and evaluated [35].

The storage of bare radioactive atomic nuclei offers the unique possibility to study the modification of nuclear half-life *e.g.* in beta decay of nuclei with empty electron shells. The discovery of the "bound beta decay", the beta decay to bound states is one of the early highlights [36]. The most recent development, is the installation of the resonant pickup at the GSI ESR. It allows the uninterrupted observation of individual atomic nuclei. For the first time an uninterrupted observation also during the cooling of recoiling daughter ions from nuclear decays opens up new possibilities for the investigation of individual nuclei [37], *e.g.* the recoil from the decay neutrino.

4. Nuclear medicine

Maria Skłodowska-Curie supported medical application of radiation. In the First World War she developed and operated the "Curie Mobile", a mobile X-ray unit to X-ray the injured soldiers at the frontline. Inspired by the benefit of X-rays she pushed the application of radium for cancer treatment. Medical application of radiation required a dose standard to guarantee a safe, which means reproducible, application of the radiation. In 1910 the dose standard: "Curie" was established. The breakthrough for Nuclear Medicine was the discovery of artificial radioactivity with ^{30}P by Irene and Frederic Joliot-Curie in 1934. In the year 1946 the routine production of radioisotopes for medical purposes started at Oak Ridge, USA.

In the recent years X-ray diagnostics and radiotherapy made large progress. A very new example is the conformal irradiation of tumors with energetic ions such beams of protons and carbon. Figure 9 shows the monitoring of a conformal cancer irradiation with a ^{12}C beam, taken with an ionization chamber placed in front of the patient. The small pictures show the slices scanning the tumor in different depths. This depth variation is

made by stepwise energy variation of the beam. The grey (green) area has already been irradiated. The inset shows an amplified example of a slice. After a project phase at GSI, a routine operation is performed at HIT, Heidelberg [38].

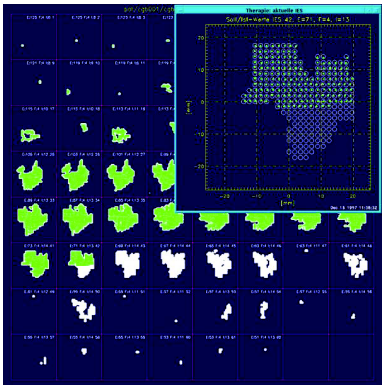


Fig. 9. Conformal irradiation of tumors: the raster scan.

5. Prospects and outlook

At present a number of "Rare Isotope" facilities for nuclear research is in operation. Next generation facilities are under way, just to mention the large-scale facilities: HIRFL-CSR at Lanzhou (China), RARF at RIKEN (Japan), SPIRAL 2 at GANIL (France), and FAIR-NUSTAR at GSI, Germany. Fig. 10 shows the FAIR facility.

Fig. 10. FAIR at GSI with the double-heavy-ion synchrotron SIS100/300, the super conducting fragment separator SuperFRS, and the storage ring system NESR, RESR/CR.

The legacy of Maria Skłodowska-Curie is not only nuclear science but also her spirit: "We must believe that we are gifted for something, and that this thing, at whatever cost, must be attained" (Maria Skłodowska-Curie).

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