FROM THE DISCOVERY OF RADIOACTIVITY TO THE PRODUCTION OF RADIOACTIVE BEAMS

R. BIMBOT
Institut de Physique Nucléaire (IN2P3/CNRS-UPS), 91406 Orsay, France

This talk is dedicated to the memory of Robert Basile (1919 - 1997), who played a major role in the development of heavy ion beams in France.

The evolution of the projectiles used to explore the nucleus influenced strongly the development of Nuclear Physics. The alpha particles from radioactivity were the projectiles mostly used up to the second world war. This period was marked by fundamental discoveries, as those of artificial radioactivity and of fission. From the 1930's to 1970, light accelerated particles (electrons, protons, deuterons, isotopes of helium) became universally used. A third period began in the 1960's with the emergence of heavy ion accelerators, the use of which led to a true revolution in the study of nuclear matter. Finally, the fourth period started in 1985 when the first secondary beams of radioactive nuclei were produced, and opened new ways in physics.

The discovery of radioactivity, by Henri Becquerel, Pierre and Marie Curie, in 1896-1898 deeply influenced the evolution of Science, Technology and Industry in the 20th century [1]. This talk will be mainly focused on the evolution of Nuclear Physics in connection with the projectiles used to explore first the atom and later the nucleus.

1. Alpha-particles from natural radioactivity used as projectiles.

One of the crucial steps in the discovery of radioactivity was achieved by Pierre and Marie Curie in 1898, when they demonstrated the presence, in Uranium minerals, of two new elements which they called Polonium and Radium. Later on, Marie Curie succeeded in isolating significant amounts of these elements, particularly of radium. Several studies had shown that the radiation from radioactivity were complex and contained three components, which were called α, β, and γ. The α component had been characterised as the emission of a heavy charged particle, which was identified later as a helium nucleus. From that period, Polonium and Radium became currently used as sources of radiation.

1.1 The Rutherford experiment (1909 - 1911)

One of the most important step in the knowledge of the atom structure was the so called Rutherford experiment realised in fact by Geiger and Marsden, in which α-particles were used to explore the structure of matter [2]. The principle of this experiment is shown in fig.1. It led Rutherford to the concept of an atom made of a central, heavy nucleus with a positive electric charge, surrounded by light, negatively charged electrons [3]. One year later, using the results of this experiment, Niels Bohr developed the planetary model of atom [4]. Beyond this very important result, the Rutherford experiment remained as a reference because it was the first time that fast particles were used as probes for studying the structure of very small objects. Experiments of the same type were performed many times with various projectiles in nuclear and particle physics, and in 1968, the discovery, at Stanford, USA, of hard structures inside the nucleons that were later identified to quarks, was the result of a similar experiment, the projectiles being GeV electrons instead of MeV α-particles [5].

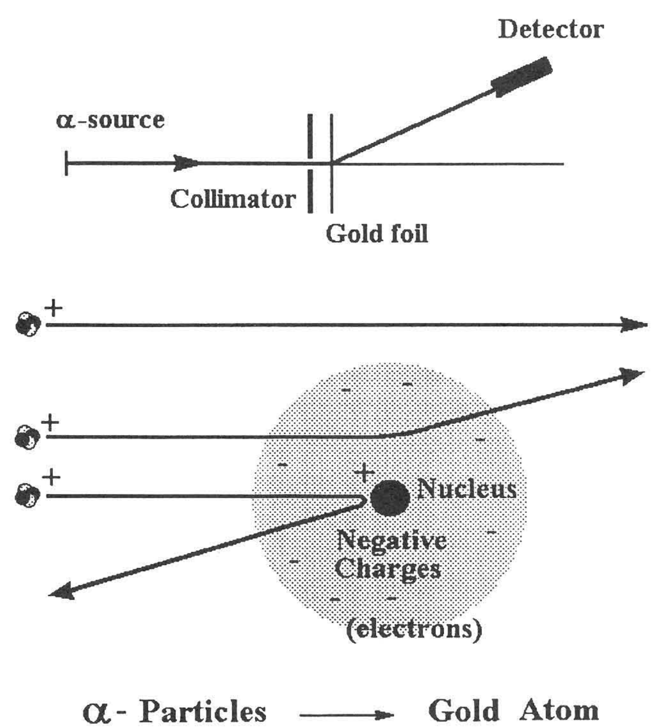

Fig 1 : The Rutherford experiment. top : principle of the experiment ; bottom : interpretation in terms of the alpha-trajectories.

1.2 The discovery of artificial radioactivity (1934)

Alpha-emitters, which provided projectiles fast enough to penetrate inside the nuclei of light elements despite the Coulomb repulsion were widely used to investigate the atom nucleus. This method led to important discoveries even after the invention of accelerators. In January 1934, four years after the invention of the cyclotron by Lawrence, the discovery of artificial radioactivity by Frédéric and Irène Joliot-Curie was thus performed using 5.3 MeV α-particles from a Polonium-210 source and an Aluminium target.

In this experiment, the two French scientists used a Geiger counter to detect the charged particles (positrons) emitted together with neutrons during the collision of α-particles and Al nuclei[6,7]. They observed that the detector went on counting when the Po source was taken away from the Al sheet, and that the number of counts per time unit was decreasing according to an exponential law, very similar to that of a radioactive decay, with a half-time of 3.25 minutes. They attributed this effect to the formation of an unknown radioactive isotope of phosphorous, ^{30}P, according to the nuclear reaction

$$^{27}Al + {}^{4}He \dashrightarrow {}^{30}P + n \qquad (1)$$

followed by the radioactive decay

$$^{30}P \dashrightarrow {}^{30}Si + e^{+} \ (+ \nu) \qquad (2)$$

where e^+ is the symbol of the positron, and ν that of the neutrino, introduced later by E. Fermi in the theory of β^+ radioactivity.

Frédéric and Irène Joliot-Curie confirmed this hypothesis by chemistry experiments in which they shown that radioactivity was following Phosphorous when this element was separated (in a few minutes!) from Aluminium.

This very important result, known as the discovery of artificial radioactivity, was at the same time the first production of a radioactive isotope and the discovery of a new mode of radioactivity, now called β^+-radioactivity. It opened the way to a systematic search for new nuclei, and to the possibility of producing artificially radioactive isotopes for medical purposes.

1.3 The neutron era and the discovery of fission (1934 - 1939)

In 1932, the neutron had been identified by James Chadwick, in experiments involving also α-particles from a Polonium source. This crucial discovery gave the key for several fundamental problems, such as the exact composition of the nucleus, the strong interaction theory (Ideki Yukawa) , the theory of β-radioactivity, and the neutrino hypothesis (Enrico Fermi). Moreover, it provided a new kind of projectile which was widely used by the Italian group of Fermi. Bearing no electric charge, the neutron is completely insensitive to the Coulomb field and can easily penetrate inside any nucleus. Taking advantage of this fact, and knowing from the Joliot's experiments that new nuclei could be produced through nuclear reactions, E. Fermi decided, in 1934 to bombard many targets with these new projectiles. Fast neutrons were mostly produced through the reaction

$$^{9}Be + \alpha \dashrightarrow {}^{12}C + n, \qquad (3)$$

similar to reaction (1), in which the α–particle was emitted by a Polonium source.

The neutrons were captured by a target nucleus ^{X}T of atomic number Z_T according to the reaction

$$^{X}T + n \dashrightarrow {}^{X+1}T + \gamma \qquad (4)$$

in which the newly produced isotope ^{X+1}T of the target element was generally radioactive, and could lead through β^- decay to a radioisotope of the following element (of atomic number Z_T+1). These radioisotopes were detected by their radioactivity, and sometimes identified with the help of chemical separations. Using this method, the Italian group produced more than 40 new isotopes in a few months [8].

Using heavier and heavier targets, they finally reached uranium in May 1934. And then, they found strange results, the capture of neutrons by uranium leading to a complicated situation in terms of radioactive half-lives of the isotopes produced. E. Fermi and his collaborators were not able to clear up this mystery, and it took three years for the European laboratories to understand that the Uranium nucleus, hit by a neutron, splitted into two components, according to a new phenomenon which was called fission. As one knows, this discovery of fission was finally achieved by the German chemists Otto Hahn and Fritz Strassmann in December 1938 and published in 1939 [9]. It had very important consequences for peaceful and military applications.

2. The cyclotron story.

2.1 Light ions machines (1930 - 1950)

As previously stated, the first concepts of particle accelerators appeared in the late 20's. Due to the general topics of this Conference, we will focus here on the development of cyclotrons.

The first cyclotron was conceived at Berkeley (USA) by Ernest Lawrence in 1929 and accelerated 80 keV protons in 1930 (fig 2). In spite of its very small size (its acceleration chamber could be taken in one hand), this apparatus was a very important realisation, and the first of an impressive series of machines of increasing power and diameter (4 inches, 11 inches, 27 inches, 37 inches, 60 inches...). In 1938, E. Lawrence was already building his sixth cyclotron! With a diameter of 184 inches (4.70m), it aimed to accelerate 100 MeV protons. This project was stopped by the war, and the magnet was used just after 1945 to build the Berkeley synchrocyclotron with which mesons were artificially produced for the first time [1].

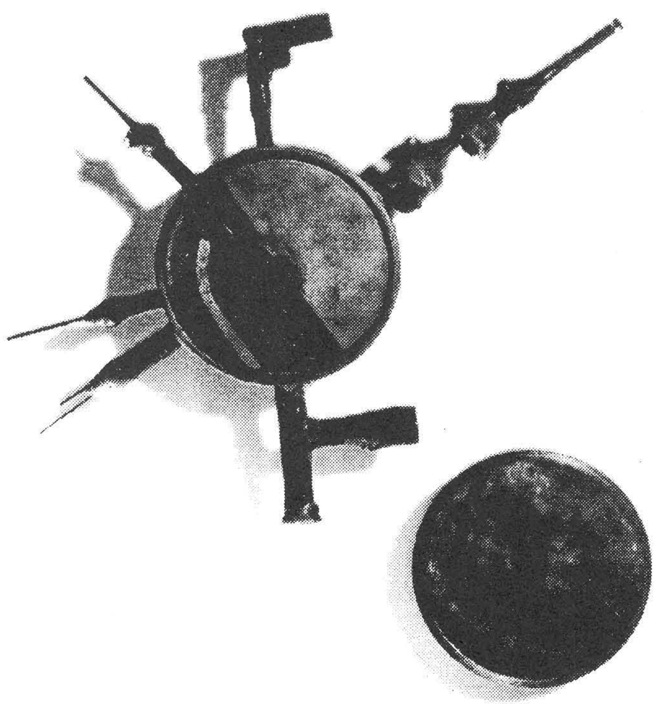

Fig 2 : The first Lawrence cyclotron. Photograph of a mock-up made at CERN. (Photo CERN).

In 1939, the first French cyclotron, which was also the first in Europe, was completed. Built by Frédéric Joliot at the « Collège de France », it had a diameter of 85 cm, and accelerated 13 MeV protons and 6.5 MeV deuterons (fig 3). This accelerator was used during many years for producing isotopes for physics and medicine, and for studying nuclear reactions. In 1957, it was transported to the Orsay laboratory, and, sixty years after being built, its magnet is still used for deviating the beam on one of the lines of the synchrocyclotron, presently used for protontherapy!

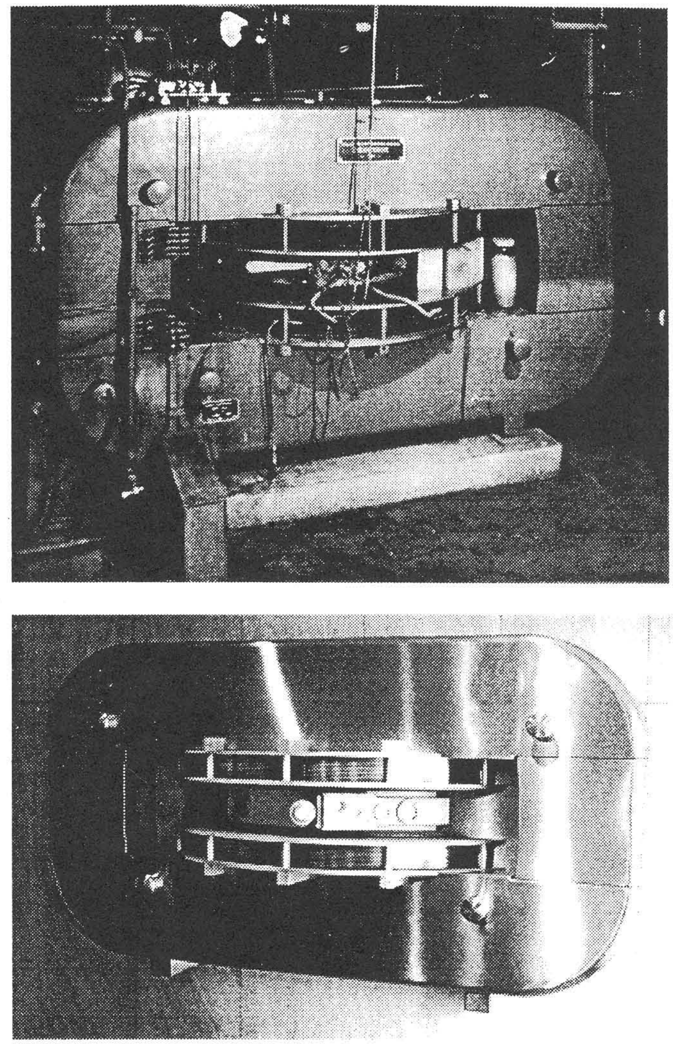

Fig 3 : Top : The 85 cm cyclotron built by F. Joliot in 1939. One can see the extracted beam.
Bottom : Evocation of this cyclotron in the IN2P3 building at the Paris CNRS headquarters.

After the second world war, powerful cyclotrons were built in several laboratories in the world. For proton projectiles, the increase in energy of conventional cyclotrons was rapidly limited by relativistic effects, and the cyclotrons had to be replaced by synchrocyclotrons. In the late fifties, in Europe, a 150 MeV proton synchrocyclotron was built at Orsay, and a 600 MeV one at CERN. At this time, High-Energy Physics separated from Nuclear Physics and became Particle Physics [10]. Powerful synchrotrons were built in several places in the world. But it was not the end for cyclotrons which still had an important role to play, in the domain of heavy ions

2.2 The first heavy ion cyclotrons (1950 - 1970)

As early as 1940, a carbon beam was accelerated to a total energy of 50 MeV in the Lawrence 37-inch cyclotron [11]. The first nuclear reactions induced by heavy ions were produced ten years later, still at Berkeley, using a 120 MeV carbon beam from the 60-inch cyclotron. In the early fifties, cyclotrons of similar sizes were used to accelerate heavy ions at Birmingham, Oak-Ridge, Saclay, Moscow and one of larger diameter at Stockholm [12].
The energy of the ions at the outer radius of a cyclotron is given by the relation

$$E = K.Q^2/A, \qquad (5)$$

in which K is a constant, which depends on the cyclotron characteristics, Q the charge state of the ion of atomic number Z (Q being smaller than or equal to Z), and A its mass. One of the main issues for increasing the energy of heavy ion beams was therefore the obtention of high charge states Q. In that purpose, multicharged ion sources were studied at the same time as the heavy ion accelerators, and in particular, the PIG (Penning Ionization Gauge) sources were developed at Dubna in the 50's [13].

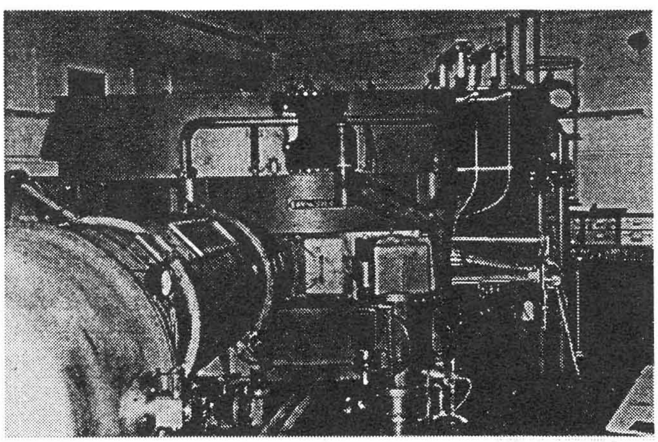

Fig 4 : The heavy ion cyclotron CEVIL built at Orsay in 1965.
(Photo IPN Orsay)

An original method had been adopted by the Birmingham engineers in order to increase the high energy component of the beam [14]. It consisted to introduce argon in the acceleration chamber. Charge exchanges occurred during the acceleration process, leading to a final beam of higher energy with a continuous energy spectrum at the outer radius of the machine. At Saclay and Stockholm, around 1955, such cyclotron internal beams were used to produce new isotopes and study heavy-ion induced reaction mechanisms [15].

Fig 5 : Robert Basile (1987)
(Photo M. Letalnet)

Heavy-ions linear accelerators (Hilac) were then built in Berkeley, Yale, Kharkov and Manchester. These machines were designed to accelerate ions up to neon at 10 MeV per nucleon using two acceleration stages separated by a gas stripper [16]. Around 1955, it was decided by Irène Joliot-Curie to build a heavy ion cyclotron in Orsay. This project had to suffer delays due to the deaths of both Irène and Frédéric Joliot, which occurred in an interval of two years (1956 and 1958). Finally, under the responsibility of Marc Lefort, the first beam from the Orsay « Variable energy heavy ion cyclotron » CEVIL [17] was delivered in 1965. The ion source of this machine had been designed by R. Basile and J.M. Lagrange [18]. This machine could only accelerate light ions, up to neon, at energies over 5 MeV/nucleon, but an extension had already been proposed by Robert Basile in 1956.

2.3 Coupled machines (1970 - 1983)

Irène Joliot-Curie had asked Robert Basile (fig 5) to study heavy ion sources for the future cyclotron, and he had the idea of using the stripping process for obtaining high charge states for projectiles much heavier than neon [19]. This method required a pre-acceleration of the ions at about 1 MeV per nucleon. After a study realised by Claude Bieth and André Cabrespine, a linear accelerator (LINAC) was built as an injector for the heavy ion cyclotron. A PIG source was producing multicharged ions for the LINAC, and a carbon stripping foil was placed at the centre of the cyclotron. The stripping conditions had been carefully studied by Eric Baron. Ions from Carbon to Krypton could thus be accelerated at energies over the Coulomb barrier of all elements up to Uranium. Typically, for Argon projectiles, Ar^{4+} ions were injected in the LINAC, and accelerated to 40 MeV. After stripping, Ar^{10+} to Ar^{13+} could be obtained and accelerated to 200-300 MeV (5-7.5 MeV per nucleon).

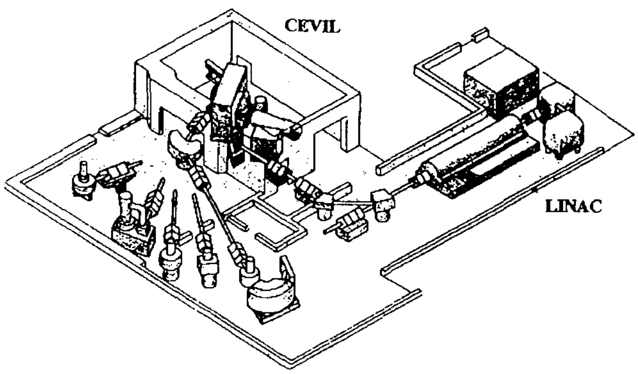

Fig 6 : Lay-out of the ALICE device, made of a linear accelerator (LINAC) injecting into the CEVIL.

This combination LINAC + CEVIL was called ALICE. This device was the first coupling of two different machines. It was also the first in the world to accelerate krypton over the Coulomb barrier of Uranium. This made it possible to search for superheavy nuclei using the fusion process (Kr + U). This research was not successful, but led to the discovery of a new reaction mechanism, which was called deeply inelastic transfer, and was very useful to study nuclear dynamics. Thus, from 1970 to 1985, the ALICE machine participated in the birth and evolution of a new and rich domain of Nuclear Physics, often called Heavy Ion Physics, in which the nucleus was considered as a new state of Matter (nuclear matter) [20]. Several projects were then developed, among which the Berkeley SuperHilac (1972), the Darmstadt Unilac (1976), and the French GANIL.

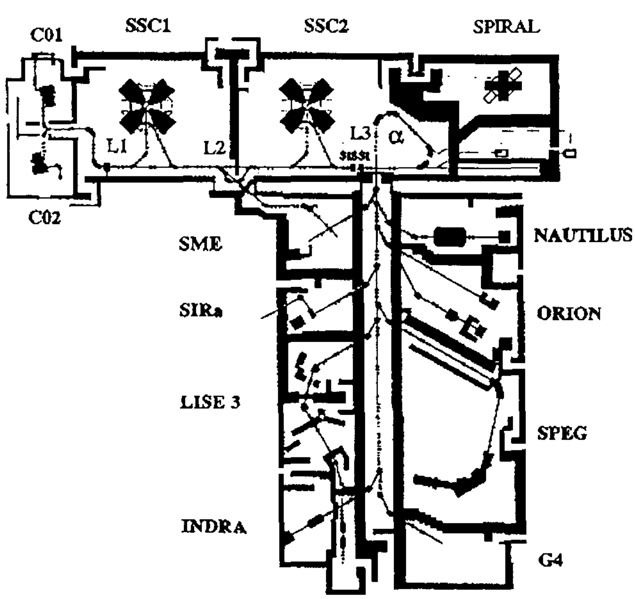

Fig 7 : Lay-out of GANIL. A compact cyclotron (CO1 or CO2) is used as an injector for two large separated sector cyclotrons (SSC1 and SSC2). An achromatic spectrometer (α) directs the beam to the experimental area. The implantation of SPIRAL is shown.

The two formers were linear accelerators able to produce beams of 10 MeV/nucleon ions of all elements up to Uranium and the GANIL device [21] was a beautiful combination of three cyclotrons (fig 7) which accelerated heavy ions from carbon to xenon in the range 20 - 100 MeV/nucleon (100 MeV/nucleon for the lightest, 20 MeV/nucleon for the heaviest). GANIL had been conceived by a national group of engineers and physicists and built under the direction of Marc Lefort and Marcel Gouttefangeas. It got its first beam in 1983. Working initially with PIG ion sources, it was modified later to take advantage of the powerful ECR sources, and accelerate all ions up to Uranium. In RIKEN (Japan), and in Lanchow (China), accelerators were also coupled to get machines working in the same energy range as GANIL.

2.4 Superconducting cyclotrons (1982 -)

Superconductivity, which had been discovered in 1911 by H. K. Onnes and interpreted theoretically in 1957 by J. Bardeen, L.N. Cooper and J.R. Schrieffer began to be applied to industrial projects in the seventies. In 1976, when it was decided to build GANIL, a choice had to be made between conventional and superconducting cyclotrons. The use of superconducting magnets made it possible to increase considerably the magnetic fields and therefore to reduce the size or increase the energy of the cyclotrons. Two superconducting cyclotrons were in construction in USA (Michigan State University) and in Canada (Chalk River). As far as GANIL was concerned, it was decided to use conventional cyclotrons, in order to get an operational machine as soon as possible. The first MSU superconducting cyclotron (K=500) was completed in 1982, and immediately, under the strong impulse of H. Blosser, the construction of a more powerful one (K=800) was undertaken. This second machine, which was finally a K=1200 machine and the real competitor of GANIL, was achieved in 1988 [22]. The K-500 and K-1200 machines are presently being coupled [23].

In Italy, a K=800 cyclotron was built in Milano and Catania and has been in operation since December 1994. This cyclotron is coupled to a tandem which acts as an injector [24].

Finally, in 1985, France and Netherlands decided to cooperate in building at Orsay a K=600 superconducting cyclotron (fig 8) which would finally be installed in Gröningen - the city where K. Onnes was born! -. This project was funded in 1987. S. Galès and H. Schreuder were in charge of its construction. It was a double challenge, firstly because it aimed at accelerating both protons and heavy ions, and secondly because it implied dismounting the machine as soon as the first beam would be obtained in order to transport it a thousand miles away! It was also a double success, the first beam being obtained at Orsay in 1994, and at Gröningen in 1995 [25].

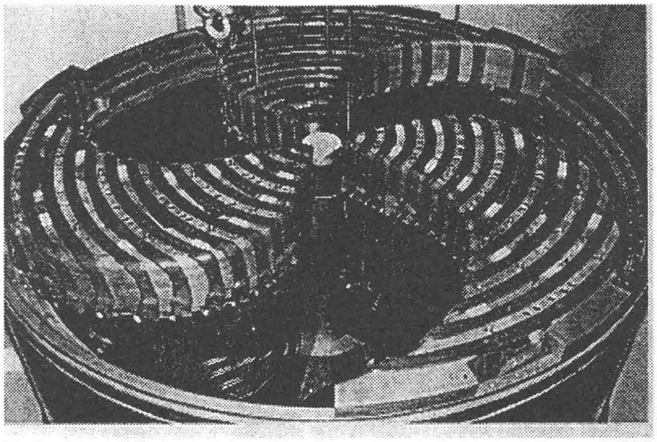

Fig 8 : One of the magnetic poles of the superconducting cyclotron AGOR (Orsay-Gröningen).

3. Radioactive beams.

An evolution similar to that induced in Nuclear Physics in the sixties with the development of heavy ion beams was going to occur twenty years later with the access to beams of radioactive isotopes. The premices of this evolution were three workshops which took place from 1981 to 1985 in the USA and in Canada. They were followed by the First International Conference on Radioactive Nuclear Beams, held in 1989 at Berkeley, 31 years after the (first) Conference on Reactions between Complex Nuclei which took place in Gatlinburg, Tennessee, in May 1958, and which may be considered as the first structuration of Heavy Ion Physics. From the experimental point of view, the most rational method for producing beams of radioactive nuclei consists in producing these nuclei at rest through a nuclear reaction induced with a primary beam in a primary target, and in accelerating them, after proper selection, to the desired energy. But historically, another method was developed first. It consisted in selecting in-flight projectile fragments from a nuclear collision, and driving them towards a detector or a secondary target. These two approaches will be presented now.

3.1 Secondary fragment beams (1979 -)

Although this technique might have been already used in a few experiments, it is in 1979 that the attention was world wide focused on the possibility of using a special beam line as a magnetic spectrometer to select and drive exotic nuclei produced through projectile fragmentation towards an identification set of detectors [26]. This experiment, realised at the Berkeley Bevalac - another interesting combination of accelerators...- using GeV/nucleon ^{48}Ca projectiles, did not use the fragment beams to induce secondary reactions, but this step was jumped over in 1985, still at Berkeley, by I. Tanihata et al [27] who measured for the first time the cross sections of reactions induced by 800 MeV/nucleon unstable nuclei.

The same year, at GANIL, the so-called LISE spectrometer began to be operated. Initially conceived for delivering ultra-stripped ions for Atomic Physics, this spectrometer had been designed by Rémy Anne to be also able to produce and separate recoil nuclei from the fragmentation of 20 to 100 MeV/nucleon projectiles [28]. Its principle is shown on fig 9. The spectrometer was originally composed of ten quadrupoles and two dipoles (1) and (2). The first dipole operated a selection of the recoiling fragments according to their magnetic rigidity. After this selection the transmitted beam was generally a mixture of different nuclei. A second selection was performed using a degrader and the second dipole, leading to a transmitted beam containing one or a few major components. The degrader had to be designed according to a well defined profile in order to preserve the system achromatism.

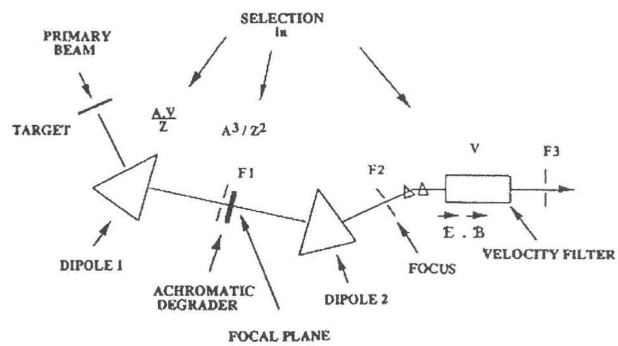

Fig 9 : Principle of the LISE spectrometer at GANIL.

In 1983, the LISE device was used as a fast collection and identification system for exotic nuclei [29]. At the same time began a systematic study of the quantitative production of radioactive fragment beams, with the objective of using them for inducing secondary reactions 30]. The first experiments of this type performed at GANIL with the LISE device were total cross section measurements. Their results were published in 1989 [31]. At the end of the 1980's, stimulated by the first results, and particularly by the discovery of neutron halo nuclei, such as ^{11}Li, the physics of radioactive beams was developing in many places. Spectrometers based on the same principle as LISE were built in several laboratories in which intermediate energy (or high energy) heavy ion beams were available, such as RIKEN in Japan, Michigan State University in U.S.A., and G.S.I. Darmstadt in Germany where a synchrotron had been coupled to the UNILAC in order to reach the energy of 2 GeV/nucleon for heavy ions. At GANIL, the LISE spectrometer was extended by an additional magnetic achromatic deviation, followed by a velocity filter, insuring the transmission of a unique secondary beam ; the two spectrometers other than LISE, i.e. the SPEG and the

ALPHA were used and/or modified to perform secondary beam experiments.

3.2 Accelerated radioactive beams (1990 -)

The in-flight selection of secondary fragments leads to the production of radioactive beams at intermediate energies while many experiments in Nuclear Physics or Nuclear Astrophysics require an incident energy close to the Coulomb barrier or even lower than the Coulomb barrier of targets. This is why, in parallel with the previous one, the second method, which implies the acceleration of radioactive nuclei produced at rest, has been developed. This method involves four main steps :

i) the production of radioactive nuclei through a nuclear reaction induced by a primary beam in a primary target. Although, in theory, the best method for this production is based on spallation reactions induced with high energy protons, - and several projects of radioactive beam accelerators are based on this principle [32] - the two examples that will be given here concern devices in which another solution has been adopted.

ii) the ionisation of the nuclei to be accelerated. The ECR ion source are presently the most efficient tools for that purpose.

iii) their selection using a more or less elaborated isotope separator,

iv) their acceleration at the required energy.

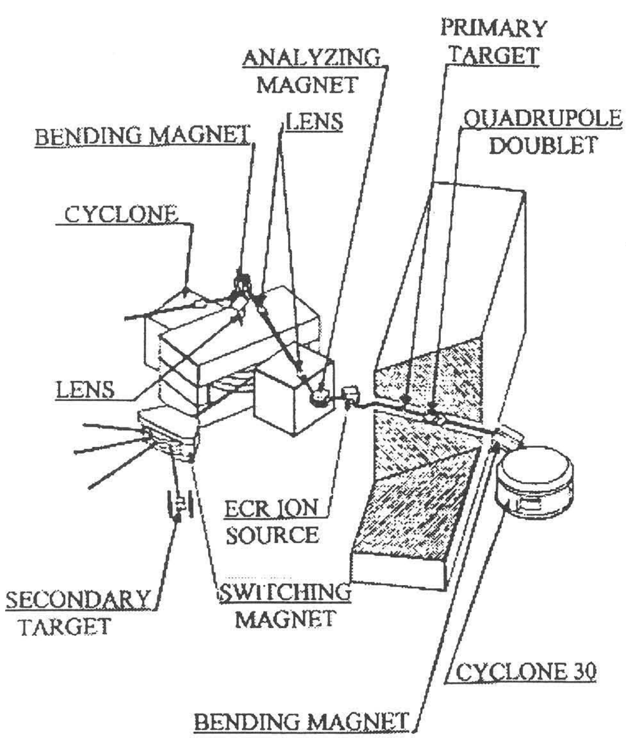

Fig 10 : Acceleration of radioactive beams at Louvain-la-Neuve (Belgium). The CYCLONE 30 cyclotron produces the radioactive species which are ionised and injected into the CYCLONE cyclotron.

The first radioactive beam ever produced through this method was obtained at Louvain-la-Neuve (Belgium) in 1990. It was a ^{13}N beam of astrophysical interest. A couple of cyclotrons were used for that purpose. The first one was a low energy (15-30 MeV) proton cyclotron (CYCLONE), delivering a 500 µA beam, which was used to produce the ^{13}N nuclei by a (p,n) reaction in a ^{13}C target. A single stage ECR ion source was used to produce $^{13}N^{1+}$ ions which were axially injected in the K=115 cyclotron CYCLONE to be accelerated to 0.65 MeV/ nucleon (see fig 10). This beam was used to measure the cross section for the nuclear reaction $^{1}H(^{13}N,\gamma)^{14}O$, of crucial interest for understanding the so-called hot CNO cycle which takes place, for example, in massive stars [33]. Since that beautiful experiment, eight radioactive species, ranging from ^{6}He to ^{35}Ar have been produced by the couple CYCLONE 30 + CYCLONE, between 0.65 and 15 MeV per nucleon [34]. A new cyclotron, called CYCLONE 44 has been recently constructed. Associated with CYCLONE, it will produce radioactive beams in the very low energy range 0.2-0.8 MeV of outstanding interest for astrophysics [35].

Finally, let us conclude this contribution by describing one of the next pieces to come on the chessboard of radioactive beam accelerators, a masterpiece called SPIRAL, which is going to deliver its first beam, 100 years after the discovery of radium! (Note that the CERN project REX-ISOLDE will be achieved at about the same time). In SPIRAL, the radioactive nuclei will be produced through the interaction of a primary beam of light or heavy ions from GANIL with a thick target. For that purpose, the intensities delivered by this accelerator have been recently improved [36}. The ECR ionisation source, especially designed, is coupled to an isotope separator which will operate a first selection of the ions before transporting them to a K= 265 compact cyclotron called CIME (fig 11). This machine will accelerate them to an energy between 1.7 and 25 MeV per nucleon.

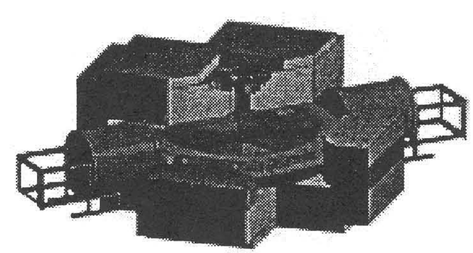

Fig11 : Computer-drawing of the CIME cyclotron, used as postaccelerator in SPIRAL (GANIL).

These radioactive beams will then be extracted and transmitted to the alpha spectrometer which will operate a last magnetic rigidity selection before directing them to the existing GANIL beam lines (see fig.7). Funded in 1993, the SPIRAL project [37] has been built at GANIL under the responsibility of Marcel Lieuvin. Technical tests of its various parts have been made during these last years, and in particular that of the CIME cyclotron which accelerated its first beam in December 1997 [38]. It was particularly moving to find, in the list of the engineers in charge of the technical design of SPIRAL, the names of Eric Baron, Claude Bieth, and Marie-Paule Bourgarel, who have participated in the French Heavy-Ion Adventure since its beginning !

I thank Eric Plagnol for careful reading of the manuscript, and Evelyne Davanture for her help in preparing this text.

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