Enrico Fermi’s Discovery of Neutron-Induced Artificial Radioactivity: A Case of “Emanation” from “Divine Providence”

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Francesco Guerra, Matteo Leone, and Nadia Robotti

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Abstract: We reconstruct Enrico Fermi's remarkable discovery of neutron-induced radioactivity in March 1934 with a focus on the experimental apparatus he used, such as the original neutron sources preserved in Italy and abroad. Special attention is paid to the role of the Radium Office of the Institute of Public Health in Rome in providing to Fermi the "radium emanation" (Radon-222) used to make his radon-beryllium neutron sources. This particular angle of investigation allows us to make a full reconstruction of what Fermi actually realized in his laboratory, to gain a better insight into his methodological choices, and, ultimately, to understand how special circumstances conspired to make the discovery of neutron-induced radioactivity possible.

* Francesco Guerra is Professor of Theoretical Physics (Emeritus) at the Department of Physics of Sapienza University of Rome. His research interests include Quantum Field Theory, Elementary Particle Theory, Statistical Mechanics of Complex Systems, and History of Nuclear Physics. Matteo Leone is Full Professor of Didactics and History of Physics at the Department of Philosophy and Educational Sciences of University of Turin. His research activities focus on the 19th and 20th centuries history of physics. Nadia Robotti is Full Professor of History of Physics at the Department of Physics of University of Genova and member of the Académie Internationale d'Histoire des Sciences. Her research activities focus on the history of atomic physics, nuclear physics and quantum mechanics.

Keywords: Enrico Fermi; Giulio Cesare Trabacchi; Radium Office; Institute of Public Health; Domus Galileana; Smithsonian Institution; artificial radioactivity; radon-beryllium neutron sources.

Introduction

This is the fourth part of a series of papers devoted to Enrico Fermi's discovery of the artificial radioactivity induced by neutron bombardment, that is, the process by which a stable material can be turned into a radioactive material by a suitable neutron bombardment. For this discovery, and for the discovery that slow neutrons increase this activity, Fermi was awarded in 1938 the Nobel Prize in Physics.

This series started in 2004 with a report about the recovery of Fermi's first laboratory notebook on this topic.[1] In June 2002, two of us (FG and NR) made an unexpected discovery in the library of a secondary technical school of a southern Italian town (the *Istituto Tecnico per Geometri "Oscar D'Agostino"* in Avellino). There, among the various documents that belonged to Oscar D'Agostino (the chemist of the so-called *Panisperna Boys,* that is, the research group—so named by Orso Mario Corbino, Director of the Institute of Physics of Rome—that gathered around Fermi and that was composed by Fermi himself, Edoardo Amaldi, Oscar D'Agostino, Emilio Segrè, Franco Rasetti, and, by September 1934, Bruno Pontecorvo), was a small dark-covered notebook. This turned out to be one of Fermi's laboratory notebooks! If that were not enough, it covered the period of March–April 1934. Both the dates and the contents handwritten on the now yellowed paper show that the notebook covers all the early Fermi's work on neutron-induced radioactivity and that it can therefore be viewed as "the first notebook of the Nobel Prize."[2] The significance of the recovery of this notebook is remarkable since Fermi never entered into much detail about the circumstances that led him to his discovery. The only information that he divulged concern his experimental apparatus and final results, eventually reported in a short letter to *La Ricerca Scientifica*, the official journal issued by CNR (the Italian National Research Council), dated March 25, 1934.[3]

This series of papers on Fermi's discovery of neutron-induced radioactivity continued in 2006 with a paper devoted to the neutron sources Fermi used, where we sought to explain how it was possible that despite the fact that others had suggested earlier that neutrons might induce such a phenomenon, it was Fermi who actually discovered neutron-induced artificial radioactivity.[4] Finally, in 2009, we carried out an analysis of the influence of Fermi's 1933 theory of beta decay on his subsequent experimental discovery.[5]

Since then, our research has investigated further aspects of the story. In particular, we have interpreted Fermi's notebook within the framework of the original instrumentation he used in his experiments, focusing on the neutron sources provided by Giulio Cesare Trabacchi, Director of the Physics Laboratory at the *Direzione Generale della Sanità Pubblica* (as the Italian National Institute of Health was then named). On the basis of this Avellino notebook, the other Fermi notebooks kept at the Domus Galilaeana in Pisa, the archival documents on the National Institute of Health preserved by the Italian State Central Archives in Rome, and the experimental apparatus that have survived, we have reconstructed all the steps Fermi made, his pace of work, and his methodological choices.

This paper focuses on Fermi's clever use of available resources, and the speed and dynamism of his progress, which opened a new route of research within only a few days. Much of the prehistory, the context, and the aftermath of the discovery is omitted, since it was covered in our previous papers, but enough is included here to understand the lessons to be learned.

Enrico Fermi, (Theoretical) Physicist

At the beginning of 1934, when he was based at the Institute of Physics of Via Panisperna in Rome, Fermi started the *experimental* research program that led him to the discovery of the artificial radioactivity induced by neutrons. Yet, a few weeks before, he had just made a great contribution to *theoretical* physics: the theory of beta decay.[6]

In December 1933, Fermi managed to explain a puzzling phenomenon, incomprehensible with current theories: without any external stimulus, some nuclei are converted into other nuclei, in a continuous way and with characteristics half-lives, with the emission of electrons with a continuous energy spectrum. To explain this, Fermi introduced a formidable idea, which would prove to be the basis of all weak interactions: the electron does not pre-exist in the nucleus, but it is created at the very moment of its expulsion together with a "neutrino," a new neutral light particle hypothesized by Pauli in 1930,[7] following the spontaneous transformation, inside the nucleus, of a neutron into a proton. In this way, the continuous spectrum of energy of the beta particles might be explained: each decay process always emits the same quantity of energy, but this energy is distributed in different ways between electron and neutrino.

Fermi's theory was symmetrical and therefore also predicted the spontaneous transformation of a proton into a neutron, with the creation, in this case, of a positive electron (positron) and an antineutrino. By this interpretation of the decay beta, Fermi discovered a new fundamental force of nature, the weak nuclear force. It is called weak because, at the energies characteristic of beta decay, which are low for the elementary particles, this force is much less intense than the strong nuclear force and the electromagnetic force. Already in his 1933 paper, Fermi estimated the value of the universal constant that characterizes the intensity of the weak interaction, now known as the Fermi constant: $g = 5 \cdot 10^{-50}$ erg $\cdot$ cm^3, a decidedly small value.[8]

The Discovery of Artificial Radioactivity

On January 15, 1934, at the *Institut du Radium* in Paris, Frédéric Joliot and Irene Curie bombarded some light elements with alpha particles and studied the effects of this bombardment using a Geiger-Müller counter, a recent instrument able to detect the presence of charged particles or other ionizing agents and to provide information about the temporal evolution of their rates.[9] They discovered an emission of positive electrons (*positrons*)[10] by the irradiated elements that continued over time, even after having removed the alpha particles source. This emission had the decreasing exponential trend typical of a radioactive element. Joliot and Curie understood that they had artificially created, through alpha bombardment, new radioactive elements that decayed by emission of β^{+} rays.[11]

A question naturally arose in Fermi's mind: since it is possible to artificially create radioactive elements that decay by β^{+} rays—that is, according to Fermi's theory, through the transformation of a proton into a neutron—is it possible to artificially perform the inverse transformation, that of a neutron transforming into a proton leading to the artificial production of radioactive substances that decay with the emission of β^{-} rays, that is, electrons?

To answer this question, again according to Fermi's theory of beta decay, the only method was to bombard the nucleus with neutrons. Once the neutron had been absorbed by the nucleus, for example through a reaction of the type (n,α), with the capture of the neutron and emission of an alpha particle, the number of neutrons inside the nucleus would increase with respect to the number of protons and with it the chance that a neutron would turn into proton.

In March 1934, three months after the formulation of his theory of beta decay, Fermi, guided by this theory, performed a difficult experiment, never attempted before. Through this experiment, on 20 March, he discovered neutron-induced radioactivity, that is the possibility of artificially creating new radioactive elements by neutrons bombardment. The experiment is conceptually the same as that performed by Joliot and Curie two months before: a sample of a substance is taken, irradiated with a neutron source, and placed nearby a suitable instrument, such as a Geiger-Müller counter, to check if the sample has become radioactive. In principle, it is a simple experiment. In practice, however, this is not the case. Why is this a difficult experiment? And how did Fermi manage to successfully perform it?

The main problem was the neutron source: whereas alpha particles are the direct product of a radioactive process and therefore are spontaneously emitted in large amounts by some natural radioactive elements, neutrons are the secondary product of a nuclear reaction that occurs at a very low rate, when a beam of alpha particles interacts with certain light elements. As a consequence, it was not possible to use neutron sources comparable in intensity to existing alpha particle sources. As Otto Robert Frisch later remarked: "I remember that my reaction and probably that of many others was that Fermi's was a silly experiment because neutrons were much fewer than alpha particles."[12]

As is well known, the neutron was discovered by James Chadwick in 1932, through alpha particle bombardment of light elements.[13] Out of several light elements, beryllium proved to be the most effective element in yielding an emission of neutrons through the following reaction:

$$ {}^{9}_{4}Be + {}^{4}_{2}\alpha \rightarrow {}^{13}_{6}C + {}^{1}_{0}n $$

The neutron sources, obtained via a secondary nuclear reaction, are in principle very weak with respect the primary alpha particle sources. For example, in the case of alpha particles emitted by polonium, the neutron rate (of a Po + Be source) is one neutron for every 30,000 alpha particles. In the case of alpha particles from radon (the Rn + Be source), the neutron rate is only marginally higher (one neutron for every 18,500 alpha particles).

Notwithstanding the higher rate of neutrons, the Rn + Be sources had several drawbacks, of which Fermi was conscious. First, radon (unlike polonium) emitted strong gamma radiation in addition to alpha particles. The neutrons obtained from a Rn + Be source were therefore accompanied by this strong gamma radiation.[14] However, the gamma-ray contamination from the Rn + Be sources was not a problem for Fermi thanks to his theory of beta decay. According to this theory, gamma rays would produce no effect on the activation of a beta radioactivity process since the presence of gamma rays would not have interfered with the possible transformation of a neutron into a proton.[15]

Another disadvantage of Rn + Be sources compared to Po + Be sources was the fact that radon ($^{222}_{86}Rn$) has a half-life of 3.82 days, much shorter than that of polonium ($^{210}_{84}Po$) which is 138.4 days. As a consequence, the Rn + Be sources had to be renewed quite often, with frequent extractions of radon from radium salts.[16] However, supply and renewal of the Rn + Be sources was not a major concern for Fermi. He could indeed rely on "divine providence," that is, on Giulio Cesare Trabacchi, Director of the Physics Laboratory of the Institute of Public Health, eventually named the "Radium Office," which was located on the basement of the Institute of Physics in Via Panisperna where Fermi operated.

Emanation from "Divine Providence"

As Laura Fermi, Enrico's widow, later recalled: "Whenever one of our physicists [Fermi and his collaborators] was in need of something, be it a screwdriver or a source of neutrons, Trabacchi was always able to provide it. The grateful young people called him nothing else but 'Divine Providence.'"[17]

Since 1923, Giulio Cesare Trabacchi (1884–1959), a former assistant of Orso Mario Corbino (Director of the Institute of Physics of the University of Rome), had been Director of the "Ufficio per le sostanze radioattive" (Office for Radioactive Substances).[*] According to

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[*] In his capacity as director of this office, Trabacchi, an accomplished experimental physicist, was involved in the tragic 1928 polar expedition of the airship *Italia*, commanded by General

the Royal Decree no. 2449, dated October 31, 1923, this office was charged with measuring natural radioactivity, chemical research on radioactive substances, preparation of radium emanation (radon), and sampling of radioactive substances.[18] On October 1, 1925, by the Royal Decree Law no. 1421, the office, which was under the Ministry of National Economy, became part of the responsibilities of the Ministry of Interior, under the "Direzione Generale della Sanità Pubblica" (which eventually became the National Institute of Health) and was charged also with "the execution of investigations for the purpose of controlling the Institutes for Physical Therapy and other physical investigations of medical interest."[19] Trabacchi's office was renamed "Laboratorio fisico della Direzione Generale della Sanità pubblica – Ufficio del Radio" (for short, the Radium Office).[20]

Independently of what the 1923 decree established, and the changes introduced by the 1925 decrees, the law in force on radioactive substances was still that enacted on December 3, 1922 (no. 1636), according to which the use of radioactive substances in medicine was controlled by the Italian state. In particular, the law stated that "the radioactive preparations owned by the State or their derivatives will be distributed … according to the availability and the respective requirements, to the university scientific institutes, with absolute precedence to those attached to the chairs of radiology and electrotherapy." Furthermore, the radioactive preparations "will also be distributed, according to convenience, to health care institutes for paid and free treatments."[21]

Umberto Nobile. Before departure, Trabacchi was asked to calibrate a Wulf electrometer to perform measurements of atmospheric electricity during the mission. Trabacchi was also asked to lend a particular type of electrometer (called Wiechert electrometer), uncommon in Italy and probably available only at Trabacchi's laboratory, which was then to be brought aboard the airship to perform delicate measurements of the electric gradient in atmosphere.

Since 1925, the Radium Office had been charged with all the technical operations connected with radium purchases and distribution to cancer treatment centers. In particular, the office calibrated all solid, liquid and gaseous preparations on behalf of those who applied to the Ministry, and prepared "radium emanation," that is, radon gas. Treatment for cancer and allied diseases used indeed either the actual radium element or the dense, inert gas, known as radium emanation or radon, extracted from radium salt solutions. This gas was purified and, by a system of mercury pumps, collected in finely drawn glass capillary tubes, sealed with a flame at both ends and cut in any desired length (figure 1). These capillary tubes containing radon were sealed either in tubes or needles, usually made of platinum, whose walls absorb most undesirable alpha and beta rays. By the early 1930s, the emanation extracted by the Radium Office in Rome was used by the Royal Physiotherapeutic Institute "S. Maria and S. Gallicano" (which, by 1933, was renamed the Royal Institute Regina Elena, as a tribute to Queen Elena). From 1926, this institute, in addition to the traditional tasks of assistance and care in the field of dermopathies, was also charged with applying physical therapies in the fight against cancer.

Fig. 1. Radon extraction plant at the Radium Office of the Direzione Generale della Sanità Pubblica in Rome. *Credit*: Archivio Fotografico del Laboratorio di Fisica dell'Istituto Superiore di Sanità, Roma

In the early 1930s, the Radium Office had two emanation plants: the first dated to 1925 and was licensed to obtain emanation from a solution containing 200 mg of radium; the second, built in 1928 entirely with materials available within the Office, exploited a solution of 1,041 mg of radium (figure 2).[22] An invoice by the Union Miniére du Haut Katanga, dated March 29, 1928, confirms the purchase of the 1041 mg sample in the form of radium hydrated bromide, at a cost of 985,306 Italian lire (approximately $52,368 in 1928 dollars) (figure 3).[23] Though this was a significant investment on the part of the Ministry of Interior, if compared with the then-annual budgets of the university-based Institutes of Physics or of the Italian National Research Council (CNR), such an investment was far more affordable than in the not so remote past. The price of the radium had dropped from over $100 per milligram immediately after World War I to about $50 per milligram in the early 1930s. This was largely the outcome of the discovery of massive rich pitchblende deposits in the Belgian Congo (now Democratic Republic of the Congo) just before the war. The introduction of Belgian radium into the world market significantly reduced the price of radium and secured a Belgian company—the above-mentioned Union Miniere du Haut Katanga—a virtual monopoly on the production and sale of radium.[24]

Fig. 2. As it can be seen in this document, by March 30, 1934, the Radium Office was in charge of 1,241 mg of radium for radon extraction purposes. *Source*: ACS, Ministero della Sanità, Istituto Superiore di Sanità, Laboratorio di Fisica – 1927–1989, box 19)

Fig. 3. The March 29, 1928, invoice by the Union Miniere du Haut Katanga for the purchase of 1,041 mg of radium by the Ministry of Interior – Direzione Generale della Sanità Pubblica *Source*: ACS, Ministero della Sanità, Istituto Superiore di Sanità, Laboratorio di Fisica – 1927–1989, box 2

The radon-filled capillary tubes usually prepared by the Radium Office for medical purposes were 40 cm in length and 0.5 mm in external diameter. Each tube was usually separated by a flame and subsequently divided into 32 pieces of about 1 cm in length (figure 4). Since the radon extraction was carried out twice a week, each piece contained about 16 millicuries, the whole tube containing about 500 millicuries.[25] However, “when special needs require it, a smaller number of more active tubes can be prepared and, where necessary, the whole emanation could be concentrated in a single tube.”[26] And special needs started occurring by late 1933, because of novel researches on radioactivity in the Institute of Physics, which shared the Via Panisperna building in Rome with the Radium Office.

Fig. 4. Snapshot from footage produced by the Istituto Luce (an Italian corporation, created during the fascist regime, involved in the production and distribution of films and documentaries) with the goal of describing the radon extraction procedures implemented by the Radium Office and the construction of the health care equipment. The snapshot shows Trabacchi’s hand separating with a flame a segment of radon-filled capillary tube. *Credit*: Archivio Storico Luce, Emanazione del Radio, 1931

Exploiting the Emanation from “Divine Providence”

At the end of 1933, Fermi secured for the first time the precious collaboration of the Radium Office for the supply of small radon sources. He and his colleague Franco Rasetti had been developing and testing a gamma-ray spectrograph. In order to develop this apparatus, they used small “glass capillary tubes, 15 mm in length and 0.2–0.3 mm in inner diameter, filled with radium emanation specially prepared … by Trabacchi” as gamma ray sources.[27]

This past experience must have influenced Fermi in the choice of a neutron source based on radon (as alpha particle source), that is, of the Rn + Be type, and must have guided it in its design and construction. In this regard Fermi, in his first letter to *La Ricerca Scientifica* (March 25, 1934) about the discovery, wrote: “The neutron source was a small glass tube containing beryllium powder and emanation. Using about 50 millicurie of emanation (which was given to me by Professor G. C. Trabacchi, to whom I extend here my cordial thanks), I could obtain more than 100,000 neutrons per second, mixed, of course, with a very intense γ-radiation; however, the latter does not influence experiments of this kind."[28]

Some time later, in the May 1934 article on the topic submitted to *Il Nuovo Cimento*, Fermi pointed out for the first time that the neutron source consisted of "a sealed glass tube about 6 mm in outer diameter and 15 mm in length,"[29] that is, the same size of the radon-filled "glass tube" previously used by Fermi and Rasetti.[30] This same type of glass tube was used to construct all the neutron sources Fermi used in subsequent experiments on neutron-induced radioactivity.

Twelve neutron sources used by Fermi and collaborators in the 1934–1938 period are currently kept at the Domus Galilaeana in Pisa, Italy (figure 5), in a wooden case (93.0 × 32.5 × 5.0 cm) wrapped in a 1.5 mm thick lead sheet (for screening purposes) (figure 6). The case contains sixteen slots carved in the wood and covered by velvet, but only twelve of them are occupied by the surviving sources (figure 7).[31]

Each source was placed at the bottom inside a long, sealed glass tube so that it could be easily and safely moved by hand. This arrangement was necessary to protect the hand by keeping it at a distance from the strong gamma radiation coming from the source, and proved particularly useful after the discovery of the neutron slowing down effect, when the various samples were irradiated by placing the neutron source inside a block of paraffin (figure 8).

These sources were donated to Domus Galilaeana in 1957 by one of Fermi's collaborators, Edoardo Amaldi. We can safely assume that there were sixteen sources at that time as the configuration of the wooden case shows.[32]

Fig. 5. Partial view of the collection of *twelve* glass tubes of different lengths (from 30 to 80 cm), containing Fermi's original neutron sources. Credit*:* Domus Galilaeana, Pisa

Fig. 6. The collection of twelve glass tubes, containing Fermi's original neutron sources, inside the lead-wrapped wooden case. *Credit:* Domus Galilaeana, Pisa

Fig. 7. Schematics of the case containing the twelve Domus Galilaeana sources, together with their respective residual radioactivity, determined on February 9–11, 2009, by the Qualified Experts in radiation protection of the Fire Department and by the Atomic Defense Laboratory of the Italian Ministry of Interior. The sources corresponding to the housings of 350, 570, 620, 630, 870 mm, have been lost. The Smithsonian source below discussed corresponds to the 350 mm housing.

Fig. 8. Schematics of the apparatus for studying the absorption of slow neutrons. The Rn + Be neutron source (S) was put inside a paraffin cylinder (P) while a second paraffin cylinder (P′) was put on P. The substance under investigation (A) was placed inside a hole excavated in the lower surface of P′. *Source*: Amaldi et al. "Artificial Radioactivity" (ref. 54), 526.

Another of Fermi's original Rn + Be neutron sources is preserved by the Modern Physics collection of the Smithsonian Institution, Washington (its accession number is 247,572 and its catalogue number in the Modern Physics collection is N-8430). As is the case for the others, this source is sealed inside a glass tube. For screening purposes, the glass tube is in turn placed inside a brass cylinder 35.3 cm long and 1.5 cm in diameter. The length of the glass tube nicely corresponds one of the empty places in the wooden case at the Domus Galilaeana. This source was donated to the Museum by another Fermi's collaborator, Emilio Segrè, in 1963. For many years it was on display in the now-closed Smithsonian exhibition "Atom Smashers," and at present it resides in storage. This source, like those preserved in Pisa (figure 7), is still slightly radioactive (figure 9).

From the correspondence kept at the archives of the Domus Galilaeana and at the Smithsonian Institution, we can reconstruct the precise trajectory of the source. Segrè wanted to donate one of the original sources to the Smithsonian, so he asked for help from Amaldi in Italy. Amaldi obtained one of the sources from the Domus Galilaeana by telling the director that they were preparing an exhibition in Rome about Fermi's discovery of neutron induced radioactivity. The donation by Segrè and Amaldi is recognized and acknowledged by official letters of Frank A. Taylor, the director pro tempore of the Museum of History and Technology at the Smithsonian, dated April 20, 1963. For tax purposes, the source was valued at $1,000. Strangely enough, there is no trace of the correspondence with the Smithsonian at the sizable Amaldi Archive at the Department of Physics of the University of Rome.[33]

Fig. 9. Fermi's neutron source preserved by the Modern Physics collection of the Smithsonian Institution, Washington. *Courtesy*: Roger Sherman, Associate Curator, Modern Physics collection at the Smithsonian

As can be seen from a laboratory notebook kept at the Domus Galilaeana in Pisa (which Fermi entitled the *Thesaurus Elementorum Radioactivorum*, as evident by the label on the cover), the sources were usually renewed on a weekly basis, always on Tuesday. The first Rn + Be neutron source was given to Fermi on Tuesday March 20, 1934, as reported on the *Thesaurus*.[34]

A Matter of Shape

If the choice of the type of source favored Fermi in his discovery, thanks to the availability of radium emanation extracted by the nearby Radium Office, Fermi's use of geometry in designing the experiment played a no less important role, both with regard to the irradiation phase and with regard to the phase of measuring any induced radioactivity.

An Rn-Be source of the type used by Fermi produces, "an isotropic [neutron] radiation in all directions."[35] To exploit this property of the source as best as he could, Fermi arranged the substances to be examined (figure 10) "in the form of small hollow cylinders" and he irradiated them by placing them "around the source." When the substances were in powder form, they were placed inside "paper containers of appropriate shape" and were irradiated with the same system.[36] These containers were colloquially called "pitali," that is chamber pots (figure 11).[37] In this way, thanks to the isotropy of the source, the various substances were subjected to neutron bombardment almost over a whole solid angle, and so the action of the source was considerably increased compared, for example, to the typical layout in which samples were placed in front of the source. Fermi would maintain this experimental layout throughout his research activity in this field.

Fig. 10. Some of the substances irradiated by Fermi. *Credit*: Museo Fermi, Rome

Fig. 11. The "pitali" containing the substances in powder form. *Credit*: Museo Fermi, Rome

Geometric factors were also crucial in the measurement phase. To determine any induced radioactivity, Fermi used a Geiger-Müller counter that he himself had designed, adopting another brilliant experimental idea. The various substances to be studied were shaped into hollow cylinders. After irradiation, in which the source was placed inside them, they were "slipped over" the counter, which was also cylindrical—naturally their respective diameters had been appropriately chosen. In this way, the losses in intensity, due to geometric factors, were "reduced to the minimum possible,"[38] since about half the solid angle could be relied on, with the result that even weak activity could be easily detected.

Counting Electrons

As Fermi reported in his May 1934 paper in *Il Nuovo Cimento*, the Geiger-Müller counters (figure 12) had centrals wires made from aluminum, following a procedure that Walter Bothe had conveyed to Bruno Rossi, one of the fathers of cosmic rays physics in Italy.[39] According to Fermi:

> The device used for detecting possible activations of the bombarded substances consisted of a Geiger-Müller wire counter. The tube of the counter was made of a thin Aluminum foil, 0.1 to 0.2 mm in thickness, such as to allow the entrance of electrons of not very large energy. The size of the counters was usually about 5 cm in length and 1.4 cm diameter. The Aluminum wire was 0.1 to 0.2 mm in diameter and was connected in the usual way to a pulse amplifier system that acted on a numerator on which the number of pulses was read from time to time. The air inside the counters was at a pressure of 5 to 10 cm of mercury, in order to have operating voltages from 1000 to 1500 volts.[40]

The numbering device Fermi used was a telephone counter of the kind used to count telephone conversations (figure 13). Since the values of the recorded counts in Fermi's Avellino notebook (for example, figure 17 below) were all four-digits numbers, we can easily deduce that the numbering device was also a four-digit one. Some examples are currently kept in the physics department of the University of Rome "La Sapienza" (figure 14).

Generally, during the periods of measurement, as the notebook shows, the Geiger-Müller counter was always left on, even in pauses during work (indeed the counts continued to grow because of the presence of background radiation). As a result, its numbering can be used by us as a sort of clock to establish the time when the various measurements recorded in the notebook were taken.

Fig. 12. One of the Geiger-Muller counters used by Fermi. *Credit*: Museo Fermi, Rome

Fig. 13. Telephone counter used by Fermi as numbering device. *Credit*: Museo Fermi, Rome

Fig. 14. Fermi's Geiger-Muller counter inserted within a counting chain (as originally displayed by the Museum of Physics of the Department of Physics, University of Rome "La Sapienza"). *Credit*: Museum of Physics, University of Rome "La Sapienza"

Amplifying Signals

Let us now make a brief reference to the amplifier Fermi used to transform the signal, produced by the passage of a beta particle in the counter, and corresponding to a voltage drop, into a current variation in the last stage capable of triggering a telephone numerator.

In the first seventeen pages of Fermi's notebook, we find the electrical diagrams of some amplification circuits for Geiger-Müller counters with the reconstruction of the characteristics of the valves employed: plate current in accordance with the grid voltage for Telefunken RE 604 and Philips A 425 and background recordings made by counters in various screening conditions and with various voltages applied.[41]

On page 4 of Fermi's notebook, we find a three-valve amplifying circuit (repeated on page 6). It is a sophisticated circuit with direct couplings between the various stages made with batteries, V_1 and V_2, without capacitors. This type of coupling had been proposed, for example by Gustav Ortner and Georg Stetter, for the amplification of transient signals, in a form called *Gleichstromverstärkung* ("direct current amplification"), which is identical to the type Fermi adopted.[42] At that time, direct coupling amplification circuits had been used in electrophysiology experiments, in which it was necessary to amplify small voltages of physiological origin without distortion. These circuits are also in use today, sought after by hi-fi enthusiasts for their effectiveness and faithfulness in the amplification of low frequencies.

From the accounting documents of the Institute of Physics of Rome, it emerges that in the previous years some "low frequency, three-stage amplifiers for photoelectric cells" had been acquired. Fermi could have adapted one of these as a signal amplifier for his counters.

The tuning of these devices is very delicate. After a few attempts, Fermi obtained "good operating conditions" for direct coupling voltages on the grids equal to $V_1 = 72$ V and $V_2 = 99$ V, respectively. In these conditions, a variation in the entry signal on the grid *G* of the first valve, from $V_3 = -9$ V to $V_3 = -4.5$ V, made the plate current of the last valve vary from I = 0 mA to I = 48 mA, sufficient to activate a telephone numbering device.

One indication of the documentary force of the notebook is that, from the calculations given on the following page (page 5), which give the operating conditions of the amplifier, both measured and calculated, one can infer that the value of the resistance r_1 through which the plate of the first valve (on the right) was powered was 500,000 Ω.

In the pages immediately following, Fermi proceeded to connect the Geiger-Müller counter, powered by high voltage, to the amplifier. From a natural interpretation of the circuit on page 15 it seems that this connection was also made directly, connecting the central wire of the counter directly to the grid of the first valve without a capacitor in between. A battery provided for the correct grid polarization. If the counter was subjected to a discharge, the grid underwent a sharp variation in potential, which was then amplified, finally activating a telephone numbering device. Fermi made other attempts to find the optimal operating conditions, also in accordance with the potential applied to the counter. These conditions finally seem to have been reached on page 17, where we find the records of various tests necessary to adjust the counter and for the measurements of background radiation.

Reducing the Background

The lower the radioactive background radiation, the stronger the statistical significance of the detected signal. To reduce the background, Fermi initially shielded the counter with bricks (figure 15). Later, he replaced them with a cylindrical lead shield partially open on one side through a removable lead door, inside which the counter was placed (figures 16–17).[43] The measurement of the possible radioactivity induced by neutrons was then determined under these setting by putting the irradiated cylindrical sample around the counter already placed in vertical position inside the lead shield, and closing the door. Details about the shielding operations are reported in Fermi's notebook (particularly in the first pages of the notebook).

Fig. 15. One of the lead bricks initially used by Fermi to reduce the background. *Credit*: Museo Fermi, Rome

Fig. 16. A vertical counter within the cylindrical lead shield preserved by the Museo Fermi. *Credit*: Museo Fermi, Rome

Fig. 17. A vertical counter within the cylindrical lead shield as depicted in a paper by Fermi (ref. 43)

The Discovery

On the night of March 20, 1934, Fermi was engaged in measurements of background radiation and in the various tests necessary to adjust the counter and amplifier (page 17). The next morning, as reported in the *Thesaurus*, he was given the first source of neutrons,[44] and began to irradiate the platinum first, without success, and then aluminum, with full success.

His actual experiments are recorded starting on page 18. After testing the counter on a sample of potassium chloride (KCl), which was known to have low beta radioactivity, Fermi, contrary to what is commonly recounted in word-of-mouth historical reconstructions, which have Fermi starting with hydrogen, chose *platinum* as the first substance to irradiate (platinum was also used as a screen for Rn + Be neutron sources against γ rays, and so it was important to ascertain that no spurious effects could arise due to any possible β activation).[45] He irradiated the platinum for 15 minutes. The counts on irradiated platinum were made at intervals of 3 minutes, 5 minutes, 10 minutes, and showed no effect.

On page 19, Fermi moved on to analyze aluminum. As a first operation, the background was measured in the presence of non-irradiated aluminum, and it proved to be about 10.5 counts per minute over a measurement of 30 minutes. He subsequently irradiated the aluminum for about 20 minutes (the irradiation time is not explicitly recorded in the notebook, but it can be estimated on the basis of the counts recorded in the meantime). The effect of the irradiated aluminum on the counter was immediately visible. Indeed, in the first 5 minutes, 82 counts were detected, which reduced in the following 5-minute intervals to 74, 59, and 57 counts, respectively. The values in the first minutes were significantly higher than background, which in the same 5-minute interval yielded about 50 counts. The results were fully compatible with β activity, provoked by neutron irradiation, which decays with a mean lifetime of about 10 minutes. A new β radioactive element had been artificially produced!

This page can rightfully be recognized as the page that records the discovery of neutron-induced radioactivity (figure 18). On this page, however, no sign, no comment, no exclamation was given. Only a red mark indicates that this measurement would be considered in the final elaboration prior to publication. It is as if nothing unexpected had occurred. Everything seemed to be predicted and everything seemed to be under control.

Fig. 18. The page of the discovery of neutron-induced radioactivity (on Aluminum) in

Fermi's notebook. *Credit*: Fondazione "Oscar D'Agostino", Istituto Francesco De Sanctis – Oscar D'Agostino, Avellino

After aluminum, Fermi was able to show neutron-induced radioactivity in Fluorine, tested in the form of calcium fluoride. At this point he decided to communicate his discovery in a letter to *La Ricerca Scientifica*. In this letter, dated March 25, Fermi proposed the following (n,α)-type nuclear reaction, based on knowledge of nuclear disintegrations with neutrons at that time, that is to say, with the capture of the neutron and subsequent emission of an alpha particle:

$$Al^{27} + n^1 \rightarrow Na^{24} + He^4,$$

where "the Na^{24} thus formed would be a new radioactive element and would be transformed into Ca^{24} with the emission of a β particle." (Fermi made a slip here: the final product of Na^{24} after a β decay is not Ca^{24} but Mg^{24}.) Fermi continued: "If these interpretations are correct, here we would have the artificial formation of radioactive elements that emit normal β particles, unlike those found by the Joliots which instead emit positrons." He concluded: "Experiments are underway to extend the examination to other elements and to study the peculiarities of the phenomenon better."[46]

As we saw above, when interpreting the results given by aluminum, Fermi assumed that the target nucleus, after capturing the neutron, emitted an alpha particle, undergoing an (n,α)-type reaction, thus being transformed into an unstable nucleus which, in turn, would undergo negative beta decay and would be transformed into a stable nucleus. The hypothesis of an initial (n,α)-type reaction, even if it was in agreement with the transmutations produced by neutrons known up to that moment, still had to be verified. The only way to do so was to chemically identify the active element that had been formed and, having identified it, to establish what reaction bound it to the starting element, thus determining if an (n,α)-type reaction, or another type, had occurred.

Fermi discovered by the chemical separation that besides (n,α)-type reactions, (n,p)-type and (n,γ)-type reactions might also occur.[47] As regards aluminum, Fermi discovered that two decays were involved: one of about ten minutes half-life and a second of about fifteen hours. Most interestingly, contrary to what Fermi wrote in his first letter to *La Ricerca Scientifica*, the ten-minute period was not due to the formation of Na^{24} by the above (n,α)-type reaction, but rather due to the formation of Mg^{27} by a (n,p)-type reaction:[48]

$$Al^{27} + n^{1} \rightarrow Mg^{27} + p^{1}.$$

Fermi determined by chemical separation that Na^{24} was actually produced, but that it was a product of the much longer fifteen-hour period.[49]

After the Discovery

As we have seen above, a few months earlier Joliot and Curie had discovered the alpha-particle-induced radioactivity in some light elements. But the neutron is much better as a projectile, since it has no electric charge and it can therefore reach nuclei more easily, even heavy ones.

As Fermi immediately showed, the neutron can create a large number of radioactive elements, throughout the entire periodic table. In this line of research, he collaborated with Oscar D'Agostino, Edoardo Amaldi, Emilio Segrè and Franco Rasetti.[50] By the summer of 1934, their efforts showed that over forty elements, out of sixty investigated, could be activated and studied with neutron bombardment.[51] This was an excellent result compared to the relatively few light elements that could be activated with charged projectiles like alpha particles, protons, and deuterons. The new radioactive elements so produced could be used for medical treatment or as tracers to follow the progress of chemical reactions and biological metabolism.[52] On May 22, 1934, Fermi gave a seminar at the Institute of Radiology of the "Policlinico" of Rome, on the medical applications of the neutron-induced radioactivity.

But a new, unexpected discovery was forthcoming. True excitement in experiment comes from unexpected results, which, when explored in detail, have dramatic implications. For Fermi, this was the discovery a paraffin moderator slows neutrons to thermal velocities and that slow neutrons have enormous cross sections and can transform virtually any element in the periodic table. In October 1934, Fermi's team, now including Bruno Pontecorvo, found that *slow* neutrons, in some cases, are much more effective in causing radioactivity compared with fast ones (such as those produced directly by the source).[23] As was later understood, nuclei indeed capture slow neutrons more easily than fast neutrons. And paraffin, or other hydrogenated substances like water,[53] are effective in slowing down neutrons when placed between the neutron source and the sample to be activated (figure 19). The effectiveness of slow neutrons was also tested on material samples that were known for not producing artificial radioactivity by fast neutron bombardment. The results with slow neutrons were surprising: these previously non-activatable materials immediately became active.

Fig. 19. One of the paraffin blocks through which Fermi discovered the effect of hydrogenated substances. *Credit*: Museo Fermi, Rome

Fermi and his collaborators reported discovery of the effect of slow neutrons in an October 22, 1934 letter to *La Ricerca Scientifica*.[54] Four days later, Fermi's team filed a patent application for this method "to increase the performance of procedures for the production of artificial radioactivity by neutron bombardment."[55] It was the triumph of the neutron, a particle capable of effectively activating the entire periodic table of the elements.

Radium versus Emanation

We have established the radium emanation from "Divine Providence" as a key factor in the discovery of neutrons-induced artificial radioactivity. This conclusion is strengthened by what occurred a few months after the discovery at the Radium Office of the Institute of Public Health. In 1934, the consensus of the radiological community was that, although the choice between actual radium and radium emanation in the treatment of tumors depends upon various circumstances, and although it was taken for granted that each method offered advantages and certain disadvantages, "at the present time there is a tendency towards the use of the actual radium rather than radon."[56]

This consensus is reflected in the results of a survey of Italian radiologists, initiated by the Direzione Generale della Sanità Pubblica on November 3, 1934, concerning the establishment of "a guiding criterion on the opportunity of varying the quantity of Radium for the extraction of the Emanation." As it was summarized by Gaetano Basile, Director of the Direzione Generale della Sanità Pubblica, in a letter to its Radium Office dated January 25, 1935, "the answers were all in agreement … relying on unquestionable facts, in considering more suitable for therapeutic purposes the use of radio element rather than the emanation." The radiologists "were unanimous in the opinion that the amount of radium in solution should be decreased." By way of example, Luigi Cappelli, vice-director of the above-mentioned Royal Istitute Regina Elena, who was critical of cancer treatments with radium emanation, while admitting that the emanation "is above all used for important physical research, especially on artificial radioactivity," proposed limiting the quantity of radium in solution for emanation extraction purposes to 500 mg.[57] As a consequence, Basile decided that since "budgetary resources do not allow … the purchase of further Radio element, in accordance with these opinions, it is necessary to use the Radium already purchased in order to have the maximum yield. To achieve this aim and to have the opportunity to answer the numerous requests received by the various cancer treatment centers, it is considered appropriate, in the meantime, to allocate the 200 mg which are in a separate solution for direct use."[58]

Contrary to the radical solutions of those who proposed sharply reducing the quantity of radium for radon-extraction purposes, Basile decided also that "the other gram of radium in solution," that used by Trabacchi for providing the radon sources to Fermi, should "still be used for the extraction of the emanation, except, at a later time, to further reduce this quantity, if circumstances recommend this."[59] It goes without saying that this decision was consequential for research at the Institute of Physics of the University of Rome, since it allowed Fermi and his group to pursue their experiments on neutron-induced artificial radioactivity. Fermi's troubles were not over, however.

In the meantime, the recently established Institute of Public Health (Royal Decree Law, January 11, 1934, n. 27) had indeed incorporated the bacteriology, chemistry, physics, biology, and malaria laboratories of the Direzione Generale della Sanità Pubblica, and notably the Radium Office, formerly housed in the same building of the Institute of Physics in via Panisperna.[60] With the establishment of the new Institute of Public Health in the building of Viale Regina Elena, the laboratories, and among them the Radium Office, had to move to the new building. In the context of the transfer of the Radium Office, on October 21, 1935, the Director of the Institute of Public Health, Domenico Marotta, addressed the issue of radon extraction in a letter to Fermi: "Having to transport the solution from which the Radio emanation is usually extracted to the new location of the Physical Laboratory, we need … to judge in good time what is the quantity of radio that must be kept in solution for the preparation of the emanation. I therefore ask Your Excellency to communicate how long you still consider that the preparations that the Physical Laboratory usually provides you with for your important research may be necessary."[61] The reasons motivating Marotta's request had to do with both scientific progress and the public health: "I am very pleased that the Institute of Health can somehow make a contribution to your work that has attracted so much interest in the scientific world in recent years and I would be very grateful if you wanted to tell me your opinion on the possibilities that radioactive substances can be used in practice to replace radium in medical applications, which would have an invaluable interest in public health."[62]

Fermi's reply was not long in coming. Two days later, on October 23, 1935, Fermi wrote to Marotta that it was absolutely desirable to continue to have significant quantities of emanation from the Radium Office (figure 20). "Research on artificial radioactivity produced by neutron bombardment has continued, as you know, for some time in this Institute; and, while encouraging results have already been achieved, many characteristics of the phenomenon, important also from the point of view of a possible production of artificial radioactive substances in considerable quantities, remain to be clarified. In order to give these researches a convenient breadth, I think it is often necessary to have, for a period of two years, quantities of 700 or 800 millicuries."[63]

Although at the beginning of the summer holidays in 1935 the working group that had formed around Fermi on the problem of neutron induced radioactivity began to break up, in the following years Fermi kept working on the study of the diffusion, slowing down, and absorption of neutrons using Trabacchi's radon sources (up to 800 millicuries in activity).[64] Still on October 17, 1938, that is, a few months before his departure for the United States, he sent a letter to *La Ricerca Scientifica* on the activation of iodine with slow neutrons, acknowledging Trabacchi's help "in providing the neutron source."[65]

As for the second issue raised by Marotta, that is, the therapeutic potentialities of artificially produced radioactive substances, Fermi was decidedly optimistic:

> On the question of what might be, in the field of medical applications, the practical significance of artificial radioactive substances, I believe I can reasonably foresee that, in the near future, it will be possible to currently produce quantities of artificial

> radioactive substances of comparable or greater activity than that of the radioactive preparations used in therapy. The artificial preparations will therefore be at least equivalent as effects, and probably cheaper, than radium. There is also the possibility, that only extensive specific research can check, that, given the great variety of elements in which radioactivity can be produced with the new methods, some of them can be found with particularly convenient chemical or physiological properties.[66]

On both grounds, Fermi's arguments proved well-founded. Concerning the quantity issue, in the near future it was indeed possible to produce large quantities of artificial radioactive substances. At first, only the cyclotron first devised by Ernest Lawrence and M. Stanley Livingstone enabled the production of radioactive isotopes in quantities sufficient for medical work; however, the nuclear reactor, the development of which was one of Fermi's main accomplishments in the United States, soon arrived to produce large quantities of these materials. Indeed, at the University of California, many of the radio isotopes formerly made in one of the cyclotrons, by 1948 were provided by one of the piles in Oak Ridge, Tennessee.[67]

As for the possibility of finding radio isotopes with particularly convenient chemical or physiological properties, it is important to recall that the possible uses of these substances are twofold: as sources of radiation in therapy and as tracers of metabolic processes. As for the latter, the case of technetium-99m is emblematic. First discovered in the University of Palermo, Sicily, in 1937 by Emilio Segrè (Fermi's former collaborator in Rome who had moved there as Professor of Physics) and Carlo Perrier (mineralogist at the University of Palermo), in a sample of molybdenum irradiated in the cyclotron at Berkeley, this technetium isotope is currently produced by thermal neutrons produced in nuclear reactors. Most importantly, in due time technetium-99m became the most commonly used radioactive tracer.

Fig. 20. Fermi to Marotta (Institute of Public Health), October 23, 1935 *Source:* Archivio Centrale dello Stato (ACS), Ministero della Sanità, Istituto Superiore di Sanità, Laboratorio di Fisica – 1927–1989, b. 5

Conclusion

The discovery of neutron-induced artificial radioactivity was accomplished with as a simple Geiger-Muller counter and a modest source of Rn + Be neutrons that easily "fits in one hand." With these modest means, Fermi accomplished what in the greatest laboratories of the world had never dared to try, thanks to his ingenuity, physical intuition, inventiveness, and tenacity, despite the limited means at his disposal.

Fermi's experimental setup nevertheless required not-so-cheap and not-so-readily available radon sources. Fermi's discovery could be made thanks to the close proximity of Divine Providence (that is, the Radium Office under Trabacchi's directorship) and, almost equally importantly, thanks to the radiological community's general dissatisfaction with the therapeutic uses of radium emanation manifesting itself when the discovery had already taken place. And, when pressure was exerted on the Institute of Public Health to reduce or eliminate the quantity of radium in solution for the extraction of radon, Marotta's foresight and Fermi's fame cooperated to allow the research of the Italian physicist to continue.

The big results achieved since then—the discovery of fission in 1938,[68] the first atomic pile in 1942, the atomic bomb,[69] the production of nuclear energy for industrial purposes, and the massive production of radioactive nuclides to be used as tracers or for medical treatment—are largely the outcome of this wonderful piece of small science.[70]

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[33] It is a pleasure for us to thank Roger Sherman, Associate Curator, Modern Physics collection at the Smithsonian National Museum of American History, for his exquisite courtesy in showing us the neutron source and the related correspondence, during a memorable and highly emotional visit at the Smithsonian, in December 2017, by two of us

(FG and NR) and Luisa Cifarelli, President of the Italian Physical Society and of the Enrico Fermi Historical Museum of Physics and Study and Research Centre, Rome.

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[37] In the copy of the volume of *Il Nuovo Cimento* that contains this article by Fermi at the Physics Department in Rome, which dates to Fermi's time, at the bottom of the page of the article where these paper containers are mentioned is a note written in pencil: "An unfortunate individual was officially assigned the difficult and boring task of constructing such containers, called in technical terms 'pitali' (chamber pots)." The conspicuously common word *pitale* is never used in the papers published by Fermi and his collaborators. Instead we find it, always accompanied by a serial number, in the laboratory notebooks or measurement cards belonging to Fermi and his collaborators to indicate some samples being examined. On the basis the handwriting, even if the author tried to modify it by using lower case block letters, it was possible to identify who wrote the phrase: it was demonstrably Enrico Fermi. We do not know who the "unfortunate individual" was. Guerra and Robotti, *The Lost Notebook of Enrico Fermi* (ref. 2), 147.

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[50] Emilio Segrè, "The Fermi School in Rome," *European Journal of Physics* **9**, no. 2 (1988), 83.

[51] Enrico Fermi, "Radioattività provocata da bombardamento di neutroni," *Ricerca Scientifica* **5**, no. 1 (1934), 330–31; Edoardo Amaldi, Oscar D'Agostino, Enrico Fermi, Franco Rasetti, and Emilio Segrè, "Radioattività 'beta' provocata da bombardamento di neutroni. III," *Ricerca Scientifica* **5**, no. 1 (1934), 452–53; "Radioattività provocata da bombarda- mento di neutroni. – IV," *Ricerca Scientifica* **5**, no. 1 (1934), 652–53; "Radioattività provocata da

bombarda- mento di neutroni. – V," *Ricerca Scientifica* **5**, no. 2 (1934), 21–22; Enrico Fermi, Edoardo Amaldi, Oscar D'Agostino, Franco Rasetti, and Emilio Segrè, "Artificial Radioactivity Produced by Neutron Bombardment," *Proceedings of the Royal Society London A* **146**, no. 857 (1934), 483–500. All are reprinted and translated in Fermi, *Collected Papers* (ref. 3).

[52] Fermi, "Prospettive di applicazioni" (ref. 43), 421–32.

[53] "It is well known that according to a widespread tradition, derived from Laura Fermi's book (ref. 17), the water in the goldfish fountain in the garden of the Physics Institute in Via Panisperna was used to produce the slowing down of the neutrons, and thus increase their efficiency in nuclear reactions. Recently the fountain was declared a Historical Site by the European Physical Society. Possible documentary evidence for this tradition can be supplied, for example, by the records on 7 November 1934 of measurements of neutron source activity, in which it is stated that the experiments were carried out in the medium 'aqua fontis,' as recorded in a laboratory notebook of Fermi's in the Domus Galilaeana.… Naturally, given the great sense of humour Fermi sometimes displayed … one might think that the appellation 'aqua fontis' chosen by Fermi simply denoted 'tap water,' compared to the solutions of boric acid H_3BO_3 in various concentrations used later. In any case, the link between the goldfish fountain and the description 'aqua fontis' is certainly appealing." Guerra and Robotti, *The Lost Notebook* (ref. 2), 231.

[54] Enrico Fermi, Edoardo Amaldi, Bruno Pontecorvo, Franco Rasetti, and Emilio Segrè, "Azione di sostanze idrogenate sulla radioattività provocata da neutroni. – I," *Ricerca Scientifica* **5**, no. 2 (1934), 282–83; reprinted in *Collected Papers* (ref. 3), 757–58; translated as "Influence of Hydrogenous Substances on the Radioactivity Produced by Neutrons. – I," in Fermi, *Collected Papers* (ref. 3), 761–62. Edoardo Amaldi, Oscar D'Agostino, Enrico Fermi, Bruno Pontecorvo, Franco Rasetti, and Emilio Segrè, "Artificial Radioactivity Produced by

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[57] ACS, 1927–1989 (ref. 22), box 5.

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[60] Regio decreto-legge 11 gennaio 1934, n. 27. Creazione e funzionamento dell'Istituto di sanità pubblica. *Gazzetta Ufficiale del Regno d'Italia*, parte prima, 27 gennaio 1934, n. 22, p. 405.

[61] ACS, 1927–1989 (ref. 22), box 5.

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[64] Edoardo Amaldi and Enrico Fermi, "On the Absorption and the Diffusion of Slow Neutrons," *Physical Review* **50**, no. 10 (1936), 899–928; reprinted in Fermi, *Collected Papers* (ref. 3), 892–942.

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[66] ACS, 1927–1989 (ref. 22), box 5.

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Museo Storico della Fisica e Centro Studi
e Ricerche Enrico Fermi
Piazza del Viminale, 1
00184 Roma RM
Italy
email: matteo.leone@unito.it