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SOME EARLY INVESTIGATIONS ON NUCLEAR ISOMERISM

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of the Artificial Radioactivity
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§ 1. Introduction. - Before the discovery of Artificial Radioactivity /1/ by I. and F. Joliot-Curie 50 years ago, little was known about the phenomenon of nuclear isomerism. Indeed such discovery gave a great impetus to the investigation of this phenomenon.

It is the purpose of this note to present a short review, covering the period until 1939, of some early investigations on nuclear isomerism. Only the very first results on each subject are being touched upon in the review. To a certain degree the report, which in no way is a full account, is given in form of personal recollections. The investigations about which quite a lot of talk is going on have been conducted at the Laboratory of Nuclear Chemistry of the College de France (directed by F. Joliot-Curie), where I was working in 1936-1939. They have been directly inspired by the discovery of Artificial Radioactivity and would not have been possible, were it not for the personal help, moral as well as material, and the never failing scientific advice of Frederic Joliot-Curie. I wish to express here my deep gratitude to this great man.

§ 2. Natural Radioactivity and Nuclear Isomerism. - The hypothesis that two atomic nuclei having the same value of the atomic number Z and the same value of the mass number A could have different radioactive properties (the hypothesis of nuclear isomerism) was put forward for the first time by Soddy /2/ in 1917. The first evidence in favour of the existence of isomers was obtained in 1921, when Hahn /3/ discovered Uranium Z. The study of the chemical and radioactive properties of Uranium Z forced Hahn to conclude that Uranium Z and Uranium X_2 are isomeric nuclei. Today it is well known that there are two beta-active isomeric forms of ^{234}Pa with decay periods 1.2 minutes and 6.7 hours, few people remembering that they were called once UX_2 and UZ.

§ 3. Artificial Radioactivity and Nuclear Isomerism. - I would like to emphasize here that for almost 20 years Uranium Z and Uranium X_2 remained the only known example of an isomeric pair. Thus nuclear isomerism for some time came to be known as an exceptional phenomenon.

After the discovery of Artificial Radioactivity, however, the search for radioactive nuclei through the bombardment of stable elements by various particles naturally led to the establishment of a number of unmistakable examples of nuclear isomerism. Especially significant and effective was the bombardment by slow neutrons /4/, which even before the advent of reactors yielded artificial radioactive nuclei all over the Mendeleyev periodic Table.

Typically, at the time (1935-1939), one could often definitely demonstrate that an isomeric pair is present, although an accurate statement about the mass number of the pair could not always been made. This, for example, happened in the case of the first discovery (1935) of an isomeric pair among artificial radioelements in bromine /5/. I shall give here one illustration; in the review article published in 1939 "Recent experimental research in nuclear isomerism" it was stated /6/ that more than thirty isomeric pairs are known to exist, whereas in the review article "Induced Radioactivity" published in the same year, only seventeen isomeric pairs with well specified properties were listed /7/.

§ 4. The discovery of Nuclear Isomerism in ^{80}Br. - The investigation of nuclear isomerism in bromine is of historical significance. As a matter of fact, the first certain proof of the existence of an isomeric pair among artificial radioactive nuclides was obtained in 1935 through the investigation of radioactive isotopes of bromine, the stable isotopes of which are ^{79}Br and ^{81}Br.

Kurchatov et al. /5/ showed that three radioactive isotopes, with periods 18 min., 4.4 hours, 36 hours are produced in the neutron bombardment of the element bromine. In a subsequent experiment of Amaldi et al. /8/ all the three periods mentioned above were shown to be sensitive to the presence of hydrogenous substances, which means that they were excited by slow neutrons. Now slow neutrons in heavy elements are simply captured. Clearly neutron capture in an heavy element consisting of two isotopes can yield three radioactive isotopes only if two periods belong to an isomeric pair.

The assignment of the mass number 80 (and not 82 !) to the isomeric pair with periods 18 min. and 4.4 hours was correctly given in 1937 by Bothe and Gentner /9/. They irradiated the element bromine with high energy (17 MeV) photons from the reaction $^{7}Li + {}^{1}H \rightarrow {}^{8}Be + \gamma$ and searched for the Br periods induced in bromine by such photons. Among the Br activities Bothe and Gentner observed the periods 18 min. and 4.4 hours, but not 36 hours, a fact which without a shade of doubt led to the assignment mentioned above: the periods 18 min. and 4.4 hours are obtained by neutron capture in ^{79}Br and by photoneutron effect in ^{81}Br.

The work initiated by Kurchatov et al. is quite typical of the first phase of research on nuclear isomerism, to which the present article is mainly dedicated. This phase, which was concluded in the early fourties with the advent of nuclear reactors and high current accelerators, was generally characterised by a relative weak activity of the artificial radioelements. Thus such weakness resulted in the circumstance that, more often than not, information on the spectra of the radiations emitted (beta, gamma and conversion electrons) had to be looked for by very rough absorption methods.

§ 5. The Weizsäcker hypothesis and early ideas on Nuclear Isomerism.- It was natural (I would say tautologically compulsory) to think that the physical difference between the two isomeric nuclei is connected with two states of different excitation of the same nucleus (say the ground and the first excited states). However one had to invoke some mechanism capable of ensuring the metastability of the excited level, that is a mechanism preserving the excited level from being destroyed very quickly by the emission of electromagnetic radiation.

As a matter of fact, experimental evidence based on the properties of the natural radioactive bodies C' (long range alpha particles) and also on our knowledge of slow neutron capture level widths (refering to highly excited levels) suggested that the radiative (γ) dipole and quadrupole transition probabilities in nuclei are of the order of $10^{12} - 10^{13}$ sec^{-1}. Now excited isomeric states may have widths smaller than 10^{-3} sec^{-1}.

The mechanism for the metastability of the excited isomeric state was proposed by Weizsäcker /10/ in 1936. He assumed that the lowest excited state of the nucleus has an angular momentum differing by several units from that of the ground state.

The Weizsäcker hypothesis played a decisive role in the early development of theoretical and experimental investigations on nuclear isomerism. I shall not be concerned in this note with explanations of nuclear isomerism different from that proposed by Weizsäcker, nor shall I touch upon the recent and important problem of spontaneous fission from excited isomeric states.

Soon after the Weizsäcker hypothesis had been formulated, in October 1937 in Paris an International Conference, very well organised by F.Joliot-Curie, took place - the Congres du Palais de la Decouverte. At the conference I put forward a few qualitative ideas, which were quite relevant, at least as far as my own subsequent work on nuclear isomerism is concerned.

Clearly of great importance in the study of nuclear isomerism was the investigation of the γ radiation emitted in the transition from the excited state to the ground state of the nucleus. However, the first searches for such radiation failed. This failure, as I suggested in 1937 /11/, might be explained if γ-rays from the excited isomeric states were strongly internally converted; in this case electrons of small energy would be emitted, which are hard to detect and had not yet been searched for.

The suggestion turned out to be right and I shall be concerned with the first experiments on nuclear isomerism and internal conversion in the next section. Here I will mention only that two theoretical quantitative papers /12,13/, based on the Weizsacker hypothesis and published in 1938, reached definite conclusions about the necessity of strong internal conversion of the low energy isomeric transition radiation.

Incidentally the (strong) internal conversion of isomeric transitions should permit sometimes to discover /11/ new isomer candidates, namely, when a very soft radiation is being observed with a period much shorter than the Sargent rules for beta decay would allow.

So far beta radioactive isomers were discussed: the isomerism in this case, implies a difference in life-times of the β-active isomers. However, as I noticed /11/ at the Congres du Palais de la Decouverte, beta-stable nuclei having a metastable excited state should also not be very rare and might be revealed by studying the radiation emitted by this metastable state. These nuclei are interesting for the understanding of nuclear isomerism, because the radiation corresponding to the isomeric transition is not troubled by the presence of unwanted beta and gamma rays. It should be possible to obtain a beta-stable nucleus in a metastable state, after a nuclear transmutation or a radioactive disintegration. However such nuclei may be obtained in a much more "clean" way, a matter which will be touched upon in a subsequent section.

At the time of the 1937 Congress it was absolutely clear to me that nuclear isomerism is by no means an exceptional phenomenon, although the actual number of known isomeric pairs was still quite small at the time.

§ 6. Nuclear Isomerism and Internal Conversion: early experiments. - In the case of beta active isomers, there is always the possibility that the radiative isomeric transition probability is decreased to such extent by the Weizsäcker mechanism, that the normal slower beta processes compete effectively to destroy the upper state. But this should not be the rule. When beta and gamma processes from the upper state have comparable probabilities and even more if the gamma process is prevailing, the radiative isomeric transition should be observable, with the reservation that, as discussed in the preceeding section, it might and often should consist mainly of (soft) conversion electrons and not of gamma rays.

From these considerations I moved when, in Paris, I initiated a search for radiative isomeric transitions. I had lot's of experience with slow neutrons. Thus good candidate targets seemed to be bromine and rhodium. From the time of the Rome work in the Fermi group I was very well acquainted with the two periods 44 sec. and 4.2 min. obtained in rhodium by slow neutrons. The 44 sec period had been used as an indicator of thermal and resonance neutrons in Rome. There I had been running cumulatively with a rhodium indicator for no less than 100 km ! It was virtually certain that the 44 sec and the 4.2 min. are isomers /14/. Thus, I selected as a target rhodium and not bromine, the reasons for the choice being rather of sentimental than scientific character. As is turned out, personally for me it was a good choice in the sense that the competition of various physicists was severe in the study of the bromine isomeric transition whereas the study of the rhodium isomeric transitions was "peaceful".

Making use of rough absorption methods and simple experimental apparatus (Rn + Be neutron sources, thin Rh targets, thin Geiger-Müller counters) I deliberately looked for a low energy electron component from Rh irradiated by slow neutrons. The soft component was actually present /15/. The experimental results could be explained by assuming that the soft radiation is an "electron line" emitted with a period 4.2 min by internal conversion in the transition from the metastable state to the ground state of the ^{104}Rh nucleus, the 44 sec. period characterising the β transition from the ground state of ^{104}Rh to the ground state of ^{104}Pd.

Similar conclusions about the strong internal conversion of the isomeric transition in ^{80}Br were independently made by Roussinow and Yuzephovitch /16/.

The results of ref. /15-16/ inasmuch as they agreed with the theoretical expectations /11-13/, based on the Weizsäcker hypothesis gave support to such hypothesis.

Already in 1939, after a number of more refined investigations had been performed, there was no longer any doubt as to the fact that isomeric transitions are often strongly converted.

First, in the cases of isomeric nuclei of radiobromine /17-18/ and of element 43 /19/, strong lines of conversion electrons have been observed in the magnetic spectrometer or in the Wilson chamber.

Second, the internal conversion is accompanied by X ray emission: as a rule the analysis of these rays is in an invaluable test in the interpretation of nuclear isomerism phenomena /19-20/.

Third, it has been possible to separate, one from the other, the two isomeric forms of radiobromine /21/. The principle of the separation method, which has been subsequently applyed to a number of other isomeric pairs, is as follows. Suppose the element, of which the isomeric states are being studied, can give compounds suitable for the application of the Szilard-Chalmers method of concentration /22/. When the isomer in the upper state decays to the lower state, the corresponding recoil may be sufficient to knock the decayed atom out of the compound. The daughter activity can then be separated, as in the classical Szilard-Chalmers method. This method has given additional confirmation that the transitions between isomeric states are strongly converted (the recoil due to the γ emission is not sufficient to knock the decayed atom out of the compound, whereas the larger recoil of a conversion electron may be sufficient).

In conclusion an additional consequence of the strong internal conversion of radiative transitions with a long lifetime (10^{-8} sec - 1 sec) should be mentioned. Such transitions should be quite frequent and therefore, as anticipated in ref. /23/, one should observe the emission of soft electron lines in the most various circumstances, in which not too light nuclei are somehow excited. Indeed this prediction has been fully verified, the first bright example being a strong component of soft electrons emitted in the slow neutron capture of gadolinium /24/.

§ 7. Beta-stable isomers. - The first example of a pair of beta-stable isomeric nuclei was discovered in the Joliot-Curie Laboratory in 1938 /25/: it was a casual discovery, but our interpretation of the phenomenon had been prepared by old and continuous thinking on the possibility (I would say the inevitability) of the existence of isomers stable from the point of view of beta decay /11/.

Having in mind problems of nuclear isomerism and internal conversion, at the time I was very much concerned with the detection of soft radiations. As a detector of soft radiation we used a cylindrical Geiger-Müller counter of effective length 40 mm. and diameter 20 mm. having an Al wall only 5 μ thick. The counter was filled with air at atmospheric pressure, so that all its area was utilized. I learned to make the counter from my friends in Florence G.Bernardini, D.Bocciarelli and G.Occhialini. Incidentally it turned out that such counters are quite capriciuous. They were not always necessary in the present experiment and in the X-ray experiment which will be described in the next section. Nevertheless they gave us at least moral help, making us certain, that we were not missing very soft radiations. I thought to use cadmium, which does not become strongly radioactive under slow neutron bombardment, as a support for a thin electrolytic deposite of the elements under study (to be activated by slow neutrons). However preliminary experiments, in which quite soft radiations could be detected, showed that, under bombardment by fast neutrons from a Rn + Be source, the cadmium cylindrical support becomes radioactive ($T \sim 50$ min), the activity being shown by chemical proofs to belong to an isotope of cadmium.

We convinced ourselved that this activity was neither produced by simple neutron capture nor by a n, 2n reaction.

We interpreted the 50 min. radiation emitted by cadmium as proceeding from a metastable state of a beta stable isotope of cadmium. The reaction of excitation without capture by fast neutrons was a familiar process to me since the old time in Rome, where the inelastic scattering of fast neutrons in lead /26/ had been investigated. The process Cd(n,nγ)Cd* was expected to have a considerable cross section (some 10^{-24}cm^2) and we thought that a part of the excited nuclei of a cadmium isotope (as it is known now, ^{111}Cd) might radiatively "fall" into the metastable state, from which the 50 min. isomeric radiation was observed.

Subsequently a similar but much cleaner case of inelastic neutron scattering to an isomer of a stable isotope of indium was studied thouroughly by Goldhaber, Hill and Szilard /27/, who moved from an old idea of Szilard. It had been known for some time that under bombardment by fast neutrons from a Rn + Be source indium becomes radioactive with a period of about 4.1 hours /26/. Goldhaber et al. confirmed and extended to other neutron sources this finding and interpreted the 4.1 hour period as a metastable isomer of ^{115}In produced in the ^{115}In(n,nγ)^{115}In*reaction, that is in a way similar to the one described above for the Cd activity /25/. The identification was certain.

First, slow neutron bombardment does not produce the 4.1 hour activity /26-27/. Second, $^{115}\mathrm{In}^*$ can grow from $^{115}\mathrm{Cd}$ in the reaction induced by fast neutrons /27/

$$^{116}\mathrm{Cd} + n \rightarrow {}^{115}\mathrm{Cd} + 2n$$
$$\hookrightarrow {}^{115}\mathrm{In}^* + e^- + \bar{\nu}$$

and from $^{114}\mathrm{Cd}$ in the reaction induced by slow neutrons /27/

$$^{114}\mathrm{Cd} + n \rightarrow {}^{115}\mathrm{Cd} + \gamma$$
$$\hookrightarrow {}^{115}\mathrm{In}^* + e^- + \bar{\nu}$$

Third, $^{115}\mathrm{In}^*$ can be produced in the reaction /28/$^{115}\mathrm{In}(p,p\gamma)^{115}\mathrm{In}^*$ induced by 5.8 MeV protons and in the reaction /29/ $^{115}\mathrm{In}(\alpha,\alpha\gamma)^{115}\mathrm{In}^*$ induced by 16 MeV α particles.

§ 8. Excitation of beta-stable isomers by X-rays. - The first experiment in which a beta-stable isomer was obtained by X-ray bombardment deserves a special place.

In the investigations which were discussed in the preceding section the metastable states of stable isotopes were generally obtained as a result of nuclear transmutations. A.Lazard and I /30/ tried a new method of producing beta-stable isomers, which makes impossible the transmutation of the nucleus and, therefore, the generation of radionuclides (the presence of which usually complicates the investigations).

The method consists in bombarding the target with a continuous X-ray spectrum of energy less than the nuclear dissociation energy. Of course, the metastable state is not excitable directly through the absorption of a quantum of energy equal to the energy of the isomeric transition. However, in a sort of nuclear fluorescence process, X-rays may excite higher nuclear levels, which combine in the spectroscopic sense of the word with the ground state, and which I shall call activation levels. The "fluorescence" gamma radiation, (usually several quanta) may leave the nucleus in a metastable state, the decay from which is observable by detecting the isomeric radiative transition.

At Ivry, in the Laboratory of Atomic Synthesis directed by F.Joliot-Curie, there was available /31/ an X-ray tube of the Brasch-Lange type, supplied by a ~3 MeV pulse generator. A rough estimate of the effective average cross section to be expected for the excitation of a few activation levels and therefore for the production of isomers by X-ray photons, showed that the facility mentioned above gave us ample opportunity to fulfil an old dream of mine: to produce isomers by X-ray photons.

Thus, using a maximum voltage of 1.8 millions volts, we looked for activities induced by X-ray bombardment in various elements.

We asked F.Joliot for some indium and got from him an In foil. We found a positive effect in indium, one of the first, if not the first, elements we investigated, the corresponding period being ~4 hours. This was obviously the $^{115}\mathrm{In}^*$ isomer referred to in the previous section /27-29/. The negative results from other elements, in which a positive effect was expected were due, as it became known later, to lack of intensity.

Our result /30/ was subsequently confirmed by M.Goldhaber et al. /32/.

At a recent neutrino conference in Sicily (1980) it was a great pleasure to meet an old acquaintance of mine, M.Goldhaber. I told him that F.Joliot, after our experiment on X-ray excitation was finished, let us know that the In foil we had been using had been sent to him by L.Szilard for some unknown reason. Maurice insured me that Szilard himself intended to perform the X-ray experiment !

Incidentally, in my work at Ivry for the first time in my life I had to deal with large scale experimental apparatus. The noise when we were measuring the generator voltage on the X-ray tube by letting sparks take place between 2 m. diameter spheres, was deafening. The Laboratory was huge, dark, very impressive and "photogenic" from the point of view of directors of films on science and magi. I liked very much to exercise in alpinism going up quickly to the top of the very high pulse generator. I remember that F.Joliot often liked to work at Ivry with his own hands.

I sent our X-ray excitation paper to my first teacher E.Fermi, who was at the time in America and had just received the Nobel Price. He sent me a letter with "heartly congratulations for the excellent results of the investigation". This was extremely gratifying; I was convinced that Fermi had some respect for me as a tennis expert (only).

F.Joliot, my second teacher, was very pleased with our X-ray results, advertised them and proposed the name of "lasting nuclear fluorescence" for the phenomenon we had discovered.

§ 9. Conclusion. - I would like to conclude my talk with the presentation of some data.

In 1934 F. and I. Joliot-Curie reported the first cases of production of artificial radioelements /1/; by 1980 there were known about 1850 artificial radionuclides (and about 280 stable nuclides).

Section 4 was concerned with the first case of nuclear isomerism among artificial radionuclides, that is with the first known artificial isomeric pair /5/; by 1980 there were known about 550 isomeric pairs (with periods > 1 sec), and about 25 "isomeric triplets", that is 25 cases where three states (with a period larger than 1 sec) are found to exist in the same nucleus.

Section 6 was dedicated to the first few cases in which an isomeric transition radiation was directly observed /15-21/; by 1980 about 250 cases were known in which the radiation from an isomeric transition (of period > 1 sec) was directly observed.

Sections 7-8 were concerned with the first examples of isomers of beta-stable nuclei and with some methods for their production /25-30/; by 1980 there were known about 50 examples of isomers of beta-stable nuclei with period > 1 sec.

This statistical material illustrates well the amazing significance of the Artificial Radioactivity discovery.

I would like to quote some words pronounced in 1936 by Enrico Fermi: "from the point of view of the history of physics the great lesson from the work of F.Joliot and I.Curie is that they have shown how it is possible to achieve great deeds with the help of simple and cheap means".

References

1. CURIE I., JOLIOT F., C.R. 198 (1934) 254.
2. SODDY F., Nature 99 (1917) 414.
3. HAHN O., Chem.Berichte 54 (1921) 1131. See also FEATHER N., BRETSCHER E., Proc.Roy.Soc.A 165 (1938) 542.
4. FERMI E., AMALDI E., PONTECORVO B., RASETTI F., SEGRE E., Ric.Scient. 1 (1934) 283.
5. KOURTCHATOW B., KOURTCHATOW I., MOUSSOWSKI L., ROUSSINOW L., C.R. 200 (1935) 1201.

6. PONTECORVO B., Nature 144 (1939) 212.
7. WALKE H. Rep.Prog.Phys. VI (1939) 16.
8. AMALDI E., d'AGOSTINO O., FERMI E., PONTECORVO B., SEGRE E., Ric.Scient. 6 (1935) 581.
9. BOTHE W., GENTNER W. Z.Phys. 106 (1937) 236.
10. WEIZSACKER C., Naturwiss., 24 (1936) 813.
11. PONTECORVO B., Travaux du Congres du Palais de la Decouverte, Paris, Octobre 1937, p. 118.
12. HEBB H., UHLENBECK G., Physica 5 (1938) 605.
13. DANCOFF S., MORRISON P., Phys.Rev., 54 (1938) 149.
14. AMALDI E., FERMI E., Phys.Rev., 56 (1936) 899; PONTECORVO B. Nature 141 (1938) 785.
15. PONTECORVO B., Phys.Rev., 54 (1938) 542.
16. ROUSSINOW L., YUZEPHOVITCH A., C.R.Acad.Sci.URSS, 20 (1938) 9.
17. ROUSSINOW L., YUZEPHOVITCH A., Dokl. AN SSSR 24 (1939) 128.
18. VALLEY G., McCREARY R., Phys.Rev., 55 (1939) 666.
19. SEGRE E., SEABORG G., Phys.Rev., 55 (1939) 808.
20. ROUSSINOW L., YUZEPHOVITCH A., Phys.Rev., 55 (1939) 979.
21. SEGRE E., HALFORD R. and SEABORG G. Phys.Rev., 55 (1939) 321.
22. SZILARD L., CHALMERS T., Nature 134 (1934) 462.
23. PONTECORVO B., C.R. 207 (1938) 230.
24. AMALDI E., RASETTI F., Ric.Scient., 10 (1939) 115.
25. DODE M., PONTECORVO B. C.R. 207 (1938) 287.
26. AMALDI E., d'AGOSTINO O., PONTECORVO B., RASETTI F., SEGRE E., Proc.Roy.Soc. A 149 (1935) 522.
27. GOLDHABER M., HILL R., SZILARD L., Phys.Rev., 55 (1939) 46.
28. BARNES S., ARADINE P., Phys.Rev., 55 (1939) 50.
29. LARK-HOROWITZ K., RISSER J., SMITH R., Phys.Rev., 55 (1939) 878.
30. PONTECORVO B., LAZARD A., C.R. 208 (1939) 99.
31. JOLIOT F., LAZARD A., SAVEL P., C.R. 201 (1935) 826.
32. COLLINS G., WALDMAN B., STUBBLEFIELD B., GOLDHABER M., Phys.Rev., 55 (1939) 507.

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Понтекорво Б. E3,4-83-581
Некоторые ранние исследования по ядерной изомерии

Представлен обзор некоторых ранних исследований /1935-1939/ о ядерной изомерии, выполненных в связи с открытием искусственной радиоактивности. Доклад не полный, он субъективный и касается только самых первых результатов по следующим темам: открытие изомерии, гипотеза Вайцекера, наблюдения изомерных переходов, внутренняя конверсия изомерных переходов, бета-стабильные изомеры и их возбуждение.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1983

Pontecorvo B. E3,4-83-581
Some Early Investigations on Nuclear Isomerism

A review is given of some early (1935-1939) investigations on nuclear isomerism conducted after the discovery of artificial radioactivity. The talk is in no way full, is subjective, and deals only with some of the very first results on the following points: discovery of isomerism, Weizsacker hypothesis, internal conversion of isomeric transitions, observation of isomeric transitions, β-stable isomers and their excitation.

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

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