On the history of study of isomerism

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Soddy's report of 1917, in which he maintained that experimental data indicated the existence of nuclear isomers, turned out to be erroneous. The first true case of nuclear isomerism was discovered by Hahn in 1921. However, the only known example of nuclear isomerism that was available for a very long time (^{234}Pa) had an extremely complicated level and transition scheme, which meant that it was impossible to establish the essence of the phenomenon. It was only after the discovery of artifical radioactivity that extensive experimental data were published and, on this basis, further research into nuclear isomerism proceeded exceedingly rapidly. Important stages in the history of research into nuclear isomerism were the identification of several cases of "an excessive number of half-lives" among artificial radioactive nuclides (In, Br, Rh), the development of theoretical descriptions of nuclear isomerism (G. Gamow, C.F. Weizsäcker, and H.A. Bethe), unambiguous demonstration of the fact that, in the case of radiobromine, the "carrier" of the two different half-lives was ^{80}Br, experiments designed to confirm the various suggested hypotheses, and the achievement of a complete understanding of the essence of nuclear isomerism. In this review, early work is discussed from the modern point of view. The importance of the work carried out by the group headed by I.V. Kurchatov on the nature of nuclear isomerism is emphasized.

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The path toward a full understanding of the physics of nuclear isomerism was, as in many other similar cases, both tortuous and long. It consisted of a gradual accumulation of initially incomprehensible facts, the emergence of a number of hypotheses for the theoretical interpretation of experimental data, the abandonment of unsuccessful hypotheses, and the acceptance of one of them on the basis of carefully designed experiments. Initially, much of the evidence was convincing but indirect. Direct evidence came later. Of course, there were some omissions and errors, too.

The history of study of nuclear isomerism is undoubtedly interesting and instructive but has not, so far, been examined in detail in the literature. Brief historical notes in papers and textbooks are often superficial and show that the authors are not familiar with primary sources.

The present sketch is intended to make good this omission, examine early work from the modern standpoint, and clarify objectively the importance of the work done by I. V. Kurchatov's group on the nature of nuclear isomerism.

Our review will be necessarily incomplete despite the fact that it is concerned with a relatively short period of time. We shall concentrate on the evolution of ideas and confine our attention to the period up to 1941, since it was during that period that the basic features of nuclear isomerism were elucidated.

1896–1921. The first decade following the discovery of radioactivity by Becquerel in 1896 was a period of very rapid progress in research into the new phenomenon all over the world. Discovery followed discovery. The work of Schmidt, Marie and Pierre Curie, Rutherfold, Debierne, Meyer, Schweidler, Fajans, and many others led to experimental and theoretical ideas that now form the basis of the science of radioactivity. Numerous members of the uranium-radium, thorium, and actinium series with strange names, such as UI, UII, AcU, RaAc, MsTh, and so on were placed within the periodic table with the aid of chemical and other methods (in some cases, incorrect assignments were made and these were subsequently corrected). It became clear that many radioactive materials had the same position in the periodic table, i.e., they were chemically indistinguishable. The idea of isotopy of chemical elements gradually emerged and was eventually rigorously formulated. In 1913, Soddy suggested that atomic species with the same position number in the periodic table but different atomic weights should be referred to as isotopes. Soon after, Moseley's research led to a new method for establishing whether particular materials with different radioactive properties were, in fact, isotopes. This was done by examining the spectrum of characteristic x-ray radiation from the material under investigation.

The first important characteristic of a given atomic species (given nuclide, in modern terminology), namely, its position in the periodic table, or the number of of charges Z, was thus determined for all the members of the radioactive families known at the time.

The situation was quite different in relation to the second important characteristic of an element, namely, its atomic weight. For example, as far back as 1902, Marie Curie found that the atomic weight of radium was 225 (she was wrong by only one unit, since it is now known that $A=226$). In many cases, especially when the lifetime was short, the atomic weight was deter-

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mined from data on parent or daughter materials, using the shift rules (Fajans, 1913). However, the use of these rules sometimes led to an accumulation of errors: if the atomic weight of a particular nuclide was determined incorrectly, the atomic weight of all the daughter nuclides was subject to the same error.1)

These errors led to a situation which we shall illustrate by only one example, although there were many similar cases of this kind: actinium-C was assigned the atomic weight of 210 and the same weight was given to radium-E; however, it was known that both nuclides were isotopes of bismuth ($Z = 83$). It was thus found that the two nuclides had not only the same position in the periodic table, but also the same atomic weight although they were in no way identical: the half-life of AcC was 2.16 min, whereas that of RaE was 5 days.2) Analysis of a number of such cases led Soddy to the following conclusion, which he reported in a paper read on May 18, 1917:[1] "Radioactive properties depend not only on the [mass of the] nucleus but also on its composition. Isotopes may exist with identical atomic weights and identical chemical character but with different stability and different decay mode. One may suppose that this much finer degree of isotopy will be found among stable elements: if this is so, it will not be possible to detect it by contemporary means.3)

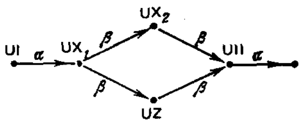
In a later lecture given on December 19, 1918,[3] Soddy gave a more detailed account of his ideas on isobaric isotopes and put forward the interesting suggestion that, if branching of a radioactive series did, in fact, result in nonidentical isobaric isotopes, this pair of nuclei would differ not only in their half-lives but also in internal energy. We note that the data on the energies of α, β, and γ transitions, which were used by Soddy as a basis for his conclusion about the energy difference between the corresponding nuclei, subsequently turned out to be erroneous and so was his suggestion that the two branches did not converge to the same nucleus. However, the idea that isobaric isotopes had different energies turned out to be correct.

As far as is known, Soddy did not undertake further specialist studies aimed at confirming the existence of nonidentical isobatic isotopes. Meyer, on the other hand, devoted considerable attention to this question. He published a detailed paper entitled, "On the existence of isotopes with equal atomic weights. Final products of the thorium disintegration series."[4] In this paper, Meyer suggests the phase, "isotopes of higher order," which was introduced by him and Schweidler at the end of 1917.[5]

Analysis of the extensive data available to him led Meyer to the conclusion that no example of this isotopy was reliably established at the time. This conclusion was repeated in a later paper.[6]

We thus see that the history of the phenomenon that was subsequently called nuclear isomerism began with a mistake: it was concluded that the phenomenon existed but it later became clear that there was no basis for this conclusion.

1921–1934. In January 1921, Hahn discovered a radioactive product belonging to the uranium series, which he called UZ.[7] This was the β-active protactinium ($Z = 91$) with a half-life of about 6.8 h. As a working hypothesis, Hahn suggested the following decay scheme for this part of the uranium series:



Soon after, Hahn published a detailed paper[8] on the properties of UZ and its position in the uranium series. He pointed out that, if his scheme were correct, one could conclude that a new type of branching, not seen before, could take place: in both branches, the disintegration proceeded by the emission of β-particles. It is interesting that Hahn did not develop this further and did not conclude that he was, in fact, dealing with a case of isotopes of higher order, which were carefully being looked for by Meyer: UZ and UX_2 had the same position in the periodic table and the same atomic weight (234), but their half-lives were different, namely, about 6.7 h and 1.14 min, respectively.

The above pair of nuclei was first noted as an example of "isotopes of higher order" in Neuburger's paper[9] (which was purely discursive and did not introduce new experimental data).

A further paper by Hahn appeared at the beginning of 1923, in which he reported his further radiochemical experiments concerned with the problem of the "uranium fork."[10] His conclusion was that it was highly probable that his "fork" was, in fact, real.

The following years saw the publication of a large number of researches designed to obtain more complete and reliable data on the details of the decay of UZ and UX_2. This was natural because this pair of nuclei was the only known example of "isotopes of higher order." Although no one was able to overturn the conclusion that these two nuclei formed an isotopic and isobaric pair, the attitude to this "knot" in the uranium series remained somewhat sceptical. The reason was that, for many years, no one put forward a hypothesis that could *explain* the fact that nuclei of the same composition had different half-lives.

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1) In its modern formulation, the "law of radioactive shifts" involves not the atomic weight (mass) but number of the nuclide.

2) In reality, the atomic weight of AcC was $A = 211$.

3) It is sometimes suggested that the first proposal for the existence of isobaric isotopes with different radioactive properties was published by Meyer and Schweidler in their monograph on radioactivity, which was widely konwn at the time[2] (p. 344). They examined the branched decay of RaC and pointed out that, after the α decay, RaC′ is transfomed into RaD, and that the product of the β-decay of RaC″ was not know. This unkown product should have been an isotope of RaD with the same atomic weight. Meyer and Schweidler continue: "The question remains whether this product is identical with RaD, i.e., whether it disintegrates in the same way as RaD into RaE and Po." Undoubtedly, Soddy succeeded in formulating this idea more clearly and more generally.

Anticipating somewhat, we note that Hahn's "fork" is, in fact, the correct scheme. However, at the time, it was represented in a different way. The complete level and transition scheme for the chain $UI \rightarrow UX_1 \rightarrow (UX_2 + UZ) \rightarrow UII$ or, in modern nomenclature, ${}^{238}U \rightarrow {}^{234}Th \rightarrow {}^{234}Pa \rightarrow {}^{234}U$, turned out to be exceedingly complicated, containing an enormous number of levels. For example, ThC′(${}^{212}Po$) has a half-life of 0.3 sec in its ground state, but it also has an isomeric state with a half-life of 46 sec.

In 1934, Gamow, who was then at the Bohr Institute in Copenhagen, published a note that contained some very important ideas on the phenomenon of "isotopy of higher order."[11] He referred to this phenomenon as "nuclear isomerism"—a briefer and subsequently generally accepted designation.[4]

Gamow's starting point was the suggestion that the nuclei might contain both protons and antiprotons but this turned out to be incorrect. It is probable that the main point of his note was not an analysis of nuclear isomerism (in the example of the "uranium fork"), but an attempt to verify the hypothesis of the antiproton. However, what he said about isomerism is the more important.

The important suggestions in Gamow's note can be formulated as follows: (1) two isomeric nuclei may differ in energy and spin; (2) when the nucleus is in a higher energy, i.e., in an excited state, it usually undergoes a transition to the ground state in a very short time (10^{-13}–10^{-15} sec) by emitting γ-rays; (3) however, in this case, this fast transition does not occur since, otherwise, one would not observe two different half-lives; one is, therefore, forced to suggest that, in the case of isomerism, one is dealing with an unusual nuclear excited state, i.e., a state with a very long lifetime, and (4) if this is accepted, one can readily explain the origin of the fact that a given nucleus may have two different half-lives.

It is almost certain that, before Gamow's paper appeared, physicists contemplating the origin of nuclear isomerism were trapped by preconceived ideas: they considered that each of the two isomeric nuclei was in its own ground state. They could not, therefore, fully understand why such nuclei had unequal half-lives, and were forced to examine the possibility that isomeric nuclei had different structure. Gamow, on the other hand, put forward his physically clear proposal, capable of resolving much of the difficulty. It was a major step forward in the understanding of the essence of nuclear isomerism. It is now known that Gamow's assumptions were, in fact, correct and are completely consistent with modern ideas on nuclear isomerism.

There is, however, a further problem that can be regarded as fundamental for the theory of nuclear isomerism. The question is: why are the lifetimes of certain excited nuclear states greater than those of the usual excited states by factors of tens or even hundreds of trillions? Gamow's answer was that, in one of the isomeric nuclei, two neutrons are replaced by a proton and an antiproton, and that the simultaneous transformation of the two particles, p and $\bar{p}$, into two neutrons (which is necessary for the transformation of one of the isomers into the other) is an event of very low probability.

Gamow's suggested existence of antiprotons in nuclei was undoubtedly regarded with great scepticism. It was not subsequently confirmed.[5] His explanation of the anomalously long lifetime of certain excited nuclear states was, therefore, incorrect.

Strange as it may seem, this error of Gamow was responsible for the fact that his other penetrating suggestions remained unnoticed or forgotten. A typical example is I.V. Kurchatov's report on new data concerning nuclear isomerism, which he gave during the discussion of I.E. Tamm's paper, "The problem of the atomic nucleus," read to a session of the Academy of Sciences of the USSR in March 1936.[12] In his concluding remarks, Tamm noted the lag between theory and experiment and stated that, "the question of nuclear isomers mentioned by I.V. Kurchatov is among those that cannot as yet be reasonably answered" (Ref. 12, p. 348). It is interesting that I.V. Kurchatov was familiar with Gamow's note; he actually mentioned it in discussion and borrowed the term "isomerism" from it.

In the course of 1934–1935, Gamow returned several times to the question of the antiproton and nuclear isomerism.[13,14] In one of his notes,[14] he used the term "metastable" to describe an excited state with an anomously long lifetime.

1934–1936. The beginning of 1934 was distinguished by an important event: I. Curie and F. Joliot discovered artificial radioactivity. They used α-particles from polonium to bombard various targets. The number of known radionuclides soon rose following the advent of different accelerators for the production of fast particles that were used to bombard targets. The number of new radionuclides rose significantly when neutrons began to be used as the bombarding particles (by Fermi's group in Rome). The eventual availability of extensive experimental data provided a new basis for researches into the various aspects of radioactivity and the properties of nuclear reactions.

The first experimental characteristic of each new radionuclide was its half-life (and, in most cases, the type and energy of the radiation associated with it). The next problem was that of "nuclide identification," i.e., the assignment of the atomic number Z and mass number A to each radioactive nucleus. There was usually little difficulty in determining Z, which was done by well-established radiochemical methods. Reliable determination of the mass number A was, how-

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[4] This designation was introduced as a clear analogy with molecular isomerism. However, according to present concepts of nuclear structure, there is no analogy between the two phenomena.

[5] The antiproton was discovered in its free state in 1955 by bombarding a target with 6.2 GeV protons accelerated in the Bevatron.

ever, much more difficult. At the time, the technology of nuclear physics experiments was in a state far removed from its present state. Natural targets that were not enriched with particular isotopes were employed, there was no direct information as to which particular nuclear reaction resulted in the formation of a particular radionuclide [for example, whether it was a (p,n) or (p,γ) reaction], and so on. The assignment of mass numbers to new radionuclides was therefore based on logical constructs based on the available knowledge of the properties of different nuclear reactions and, frequently, with the aid of the very effective method of "crossed nuclear reactions."

Soon after the onset of extensive studies of artificial radionuclides, unexplained situations arose which could be referred to as the "problem of the excessive number of half-lives." The first case of this kind was described by Szilard and Chalmers.[15] It had been known before this note appeared that two radioactive isotopes of indium were produced in the $(\mathrm{In}+\mathrm{n})$ reaction with half-lives of 12 sec and 54 min, respectively, and that their intensity increased sharply as the neutrons were slowed down. A third half-life of about 3.5 h was reported in Ref. 15. On the other hand, mass spectrometric data showed that indium had two stable isotopes and the abundance of one of them, namely, ^{115}In, in natural indium was greater by a factor of 10 than the abundance of the other (^{113}In). Slow neutrons could produce only the (n,γ) reaction, so that the two radionuclides, ^{114}In and ^{116}In, could be produced by exposing natural indium to slow neutrons. The question was—which nuclide had the half-life of 3.5 h? Szilard and Chalmers assumed that this was the $Z=49$ isotope of indium, on the basis that reactions such as $\mathrm{In}(n,\alpha)$ and $\mathrm{In}(n,p)$, which could lead to nuclides with other values of Z, had never been observed in targets heavier than zinc ($Z=30$) when neutrons from the $(\mathrm{Ra}+\mathrm{Be})$ source were employed.

But they did not perform an experiment capable of showing whether the activity of the new nuclide was enhanced when the target was surrounded with water (i.e., when the neutrons were slowed down).[6]

Szilard and Chalmers noted that the discovery of the third half-life in indium bombarded with neutrons gave rise to a situation that could not be readily understood. This case was said to "deserve further investigation, for which adequate instruments of observation are not at present at our disposal." The question was thus formulated, but Szilard and Chalmers put forward neither a solution nor any hypothesis.

It is now known that In (13 sec) is, in fact, ^{116}In and In (54 min) is ^{116m}In (the superscript m indicates a metastable state). The half-life of 3.5 h was subsequently found to be 4.5 h and was assigned to ^{115m}In which was produced in the $^{115}\mathrm{In}\,(n,n')$ reaction. Thus, Szilard and Chalmers, who performed their work in 1934, in fact observed two isomeric pairs of nuclei, but were not able to verify this point.

None of the researchers discussing the puzzle of the "excessive number of half-lives" suggested, at the time, that the list of nuclear reactions that could have been responsible for the appearance of a particular radionuclide should have included inelastic scattering of neutrons or charged particles, i.e., reactions of the form (n,n'), (p,p'), and so on. This is not a surprising omission since we are dealing with the very earliest stage of research into the properties of artificial radionuclides.

Had Szilard and Chalmers used better equipment and bombarded indium with neutrons for a longer time, they would have found that the chemical residue corresponding to indium contained two more radionuclides, with half-lives of 50 days and 99.4 min, respectively. One of them would have been ^{114m}In, resulting from the reaction $^{113}\mathrm{In}\,(n,\gamma)$ and the other, ^{113m}In, originating in the $^{113}\mathrm{In}\,(n,n')$ reaction.[7]

It is not difficult to imagine that discovery of five radioisotopes of indium after the bombardment of indium with neutrons would have faced the researchers with a dilemma that would have been much more profound than the discovery of the three isotopes.

The second puzzle, relating to the "excessive number of half-lives," emerged after the paper by B.V. Kurchatov, I.V. Kurchatov, L.V. Mysovskii, and L.I. Rusinov[18] and the papers that soon followed it.[8] They studied the γ-ray activity of radionuclides produced when bromine was irradiated with neutrons from an $(\mathrm{Ra}+\mathrm{Be})$ neutron source, and discovered three rarionuclides with periods of 18 min, 4.5 h, and 36 h. The first two had already been seen by the Fermi group, who initially reported half-lives of about 30 min and 6 h, and then 18 min and 4.2 h.[19,16] The Rome group showed that these two nuclides were isotopes of bromine and had a high "coefficient of sensitivity to water." Since bromine consisted of two isotopes (^{79}Br and ^{81}Br, in roughly equal amounts), it was assumed[19] that these half-lives could be assigned to ^{80}Br and ^{82}Br—of course, without indicating which period was due to which particular isotope, since there was no evidence available for drawing such a conclusion. There was, however, no doubt as to the fact that it was the (n,γ) reaction that was responsible for the appearance of these two nuclides. I.V. Kurchatov *et al.* used chemical data to

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[6] Fermi's group published a review paper dealing with the radionuclides produced when the targets were irradiated with neutrons[16] a few months after the paper by Szilard and Chalmers.[15] They noted that the nuclide with a half-life of about 3 h was identified radiochemically as an isotope of indium and that it was practically insensitive to the slowing down of neutrons.

[7] The $^{113}\mathrm{In}\,(n,\gamma)$ reaction would have resulted in a further radioisotope of indium, namely, ^{114}In (72 sec) but, for well known reasons, the corresponding cross section is exceedingly small,[17] so that it is practically unobservable.

[8] I. V. Kurchatov was the Director of the Laboratory for the Atomic Nucleus at the Leningrad Physicotechnical Institute and L. I. Rusinov was a member of the staff of this laboratory. The chemist, B. V. Kurchatov, worked in the same institute. L. V. Mysovskii was the Head of the Physics Department of the State Radium Institute in Leningrad.

show that the 36-h radionuclide was an isotope of bromine. The two stable isotopes of bromine produced three active isotopes under neutron irradiation, two of which must have been ^{80}Br and ^{82}Br. The question was—what was the mass number of the third bromine isotope? In an attempt to answer this question, Kurchatov *et al.* listed the following possibilities: (1) it may be that, under the influence of the neutron, the bromine nucleus emits simultaneously two particles, one of which is positive and the other negative (it appears that they had a proton and an electron in mind, in which case the result should have been ^{78}Br), and (2) isomeric nuclei may have been produced. Kurchatov *et al.* concluded, however, that these two hypotheses had to be "excluded as too special."

It is clear that the first of the two hypotheses was an attempt, in a difficult and puzzling situation, to explain one new phenomenon in terms of another that had not been observed. As regards the hypothesis about the existence of isomeric nuclei, we note that this was meant to be Gamow-type isomerism based on the assumed existence of antiprotons as components of the nucleus. Kurchatov *et al.* could not bring themselves to take this seriously.

Nevertheless, this first mention that the puzzle of "excessive of half-lives" could, in principle, have been solved in terms of the idea of isomeric nuclei is of major importance, especially since it was published in a widely known journal.

Having rejected the above two hypotheses, Kurchatov *et al.* considered a third: it was possible that ^{78}Br, with a half-life of 36 h, was produced in the $(n,2n)$ reaction.[9)] Their paper ends with the remark: "to verify this hypothesis, we will have to investigate the reaction yield as a function of neutron energy." In fact, high-energy neutrons (about 10 MeV) must be used to produce the $(n,2n)$ reaction.

In accordance with the program outlined in Ref. 18, L. I. Rusinov proceeded to determine the yield of Br (36 h) under bombardment with neutrons from different sources, including (Ra + Be), (Ra + B), (Ra + Li), and so on.[20] These experiments clearly demonstrated that the reaction responsible for the production of Br (36 h) did not have an energy threshold and there was a large "coefficient of sensitivity to water." It became clear that the third radiobromine was produced in the (n,γ) reaction.

The Fermi group in Rome arrived at the same conclusion independently. Having become familiar with the work of Leningrad physicists,[18] they immediately proceeded to carry out the necessary experiments.[21] They confirmed that the Br (36 h) was produced and showed that all three radiobromines had a high "coefficient of sensitivity to water." The hypothesis involving the $(n,2n)$ reaction could not, therefore, be adopted. The authors noted that the problem of the third radiobromine was very interesting, and that possibly the situation may be similar to that in the case of indium.[15] However, no solution was proposed.

In their paper, "An investigation of natural radioactivity under neutron bombardment. Part I," submitted toward the end of 1935, Kurchatov *et al.*[22] put forward a clear formulation of the problem of "excessive number of halflives." They wrote: "Detailed studies of artificial radioactivity have only just begun and there is a number of unexplained features that are of great interest. They include, for example, ...the nature of radioisotopes of elements for which the number of observed half-lives is greater than the number of isotopes and for which chemical analysis shows that radioactive nuclei are isotopes of the irradiated element." They gave bromine as an example.

Before we examine further the history of radiobromine, let us consider another early case of "excessive number of half-lives." When rhodium is exposed to slow neutrons, two radionuclides are produced, namely, rhodium isotopes with periods of about 50 sec and about 5 min[19] (these values were subsequently improved to 44 sec and 4.7 min, respectively). It was shown that these two isotopes had a high "coefficient of sensitivity to water."[16] On the other hand, it was firmly established that natural rhodium had only one stable isotope, namely, ^{103}Rh. Thus, the case of rhodium is the clearest and most obvious example of the "excessive number of half-lives": both half-lives have to be assigned to the same nucleus (^{104}Rh) produced in the (n,γ) reaction. Nevertheless, this fact was not commented upon, nor was it noted that it required an explanation. On the other hand, Kurchatov *et al.*[22] did note that rhodium and bromine were, in that sense, similar. Much later, it was noted in one of the reveiw papers by the Fermi group[23] (at the end of 1936) that three cases—Rh, In, and Br—exhibited an unexplained excessive number of half-lives.

It is now known that Rh (44 sec) is ^{104}Rh and Rh (4.7 min) is ^{104m}Rh (where the metastable state is not the first but the third excited state of ^{104}Rh). Finally, a further radioisotope of rhodium, namely, ^{103m}Rh (57 min), is produced in the ^{103}Rh (n,n') reaction when unmoderated neutrons bombard rhodium. This was not found in Ref. 23.

Having had at his disposal the new data showing that Br (36 h) was produced in the (n,γ) reaction, I. V. Kurchatov returned, toward the end of the same year (1935), to the hypothesis of nuclear isomerism, which he rejected at the beginning of that year.[24] He gave a most complete exposition of the essence of the problem of radiobromine together logical with arguments excluding a number of hypotheses. He did this in two public lectures, namely, a lecture read at the N. D. Zelinskii PhysicoChemical and Power University[25] (probably toward the end of 1935 or beginning of 1936) and in the discussion following Tamm's paper at the March session of the Academy of Sciences of the USSR in 1936, mentioned above.[26] After a critical examination of a number of hypotheses. Kurchatov noted that "we must

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[9)] The possibility of the $(n,2n)$ reaction had already been mentioned in the early work by Fermi's group, for example, in Ref. 19 (p. 497). However, this reaction had not been detected experimentally at the time.

conclude that the mass number and the position of an element in the periodic table do not define all the properties of the nucleus.... It would appear that one must admit the possibility of isomeric nuclei; these are *two isotopes with the same mass number but different structure*" (Kurchatov's italics).[25] Kurchatov also pointed out, during the March session of the Academy of Sciences, that the observed data could be explained by admitting the existence of isomeric nuclei. He added: "When I speak of nuclear isomerism, I do not wish to treat it in the sense put forward by Gamow." (He then repeated the definition of nuclear isomerism.)

We thus see that Kurchatov appears to have corrected the logical inconsistency in Ref. 18 in which he rejected the hypothesis of isomerism as a solution to the radiobromine puzzle. Having reviewed in detail the then available experimental data, he came to understand that, although his rejection of the Gamow nuclear isomerism was correct, this did not necessarily mean that the very concept of nuclear isomerism (as defined by Soddy) had to be rejected.

Two questions, both elementary and unrelated to the attempts to establish the nature of nuclear isomerism, remained open: which particular bromine nucleus exists in the two isomeric forms, and which two out of the three half-lives can be assigned to them? The answer came later.

In order to resolve the puzzle of the third radiobromine, Blewett undertook a careful investigation of the isotopic composition of bromine.[27] He confirmed with considerable precision the results that had already been known, namely, that there are two stable bromine isotopes in nature ($A = 79$ and 81). For the hypothetical third stable isotope of bromine ($A = 77$ or 83), he established an upper limit: its relative concentration in bromine does not exceed one part in 3000. It follows that, if this rather rare isotope is transformed into one of the three radioactive isotopes of bromine as a result of the (n, γ) reaction, the cross section for this reaction must be greater by a factor of 500 than for either of the other two remaining bromine isotopes. It is clear that this explanation of the radiobromine puzzle had to be rejected.

It is known that, when bromine is bombarded by neutrons that have not been slowed down, a further radioisotope of bromine, namely, ^{79m}Br (~4.9 sec) is produced in the ^{79}Br (n, n') reaction. As in other similar cases, the fact that this radionuclide was unnoticed in 1935–1936 was actually favorable since, otherwise, the identification of all the half-lives of bromine would have turned out to be even more difficult.

The end of 1936 was distinguished by an important event in the history of nuclear isomerism. Weizäcker published his paper, "Metastable states of atomic nuclei,"[28] which was the culmination of the work he began at Bohr's Institute in Copenhagen. In this paper, he finally put forward a satisfactory theory or, more precisely, hypothesis for the origin of nuclear isomerism. Comparison of the relevant dates will make it clear that Weizäcker was not familiar with Kurchatov's remarks made at the March session of the Academy of Sciences. It appears that Lise Meitner drew his attention to a few cases among artificial radionuclides which could have been examples of nuclear isomerism. In the summer of 1936, she noted,[29] at a seminar held in Zürich, that several incomprehensible cases, for example, Br, In, Rh, and U, could have been explained by assuming (albeit reluctantly) the existence of nuclear isomers. Weizäcker refers to Ref. 29 and at the end of his paper thanks Lise Meitner for useful discussions.[10)]

Weizäcker does not refer to Gamow's paper. However, he reproduces precisely the same assumptions: one of the members of the isomeric pair must be a metastable excited state of the nucleus, whereas the other member of the pair must be a nucleus of the same composition but in the ground state. There follows the new and basic idea of this paper: the reason for metastability is a strong quantum-mechanical selection rule which substantially reduces the probability of a γ-transition from the excited to the ground state, i.e., the selection rule that results in a long lifetime of the metastable nuclear excited state. This is associated with two facts, namely, the relatively low excitation energy of the metastable state and the relatively great difference between the angular momenta (spins) of the metastable and ground states. The multipole order of the emitted γ-radiation depends on this spin difference ΔI. When dipole radiation is emitted ($\Delta I = 1$), the transition probability is very high; when quadrupole radiation is emitted ($\Delta I = 2$), the probability is much smaller, and so on. Weizäcker reproduces a very approximate formula for the lower limit of the lifetime τ_γ of the excited nuclear state for the γ-transition to the ground state. This formula predicts that, for example, for a nucleus with $Z = 27$, excitation energy 50 keV, and $\Delta I = 4$, the lifetime of this excited state is $\tau_\gamma \geq 100$ sec.

Weizäcker notes that, in general, the transition from the excited to the ground state occurs not only via the emission of a γ ray but, at the same time, by the emission of internal conversion electrons. He then points out that Niels Bohr drew his attention to certain general properties characterizing the motion of nuclear matter, which ensure that the actual lifetime of a metastable state for multipolar γ-transitions may be even greater than the value predicted by the simple formula.

It was known that a nucleus occupying an ordinary excited state does not decay by the emission of β-particles because there is a much more probable and faster mode, namely, γ-transition. However, in the case of metastable states, the situation is different and Weizäcker assumed that the nucleus would decay from this type of level by emitting a β-particle. Weizäcker's ideas can be illustrated by the scheme shown in Fig. 1, which gives the simplest possible transition modes. The number of γ-transitions shown in this figure is, of course, very tentative. There is one further possible

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[10)] It is quite possible that the phrase, "isomeric nuclei" and, perhaps, even the idea of isomerism as a way of explaining incomperhensible facts, were borrowed by Meitner from the paper by Kurchatov *et al*.[18]

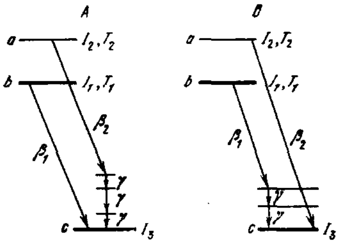

FIG. 1. Decay scheme of an isomeric nucleus (according to Weizsäcker): a—metastable state, b—ground state of the isomeric nucleus (the alternative terminology is: states a and b are isomeric nuclei or an isomeric pair). $|I_2 - I_1| \geq 2$. In general, the half-lives T_1 and T_2 for the β-transitions are different. This is, in fact, observed in the case of nuclear isomerism.

variant which, for simplicity, is not shown in Fig. 1 and in which none of the β-transitions proceeds to the ground state c. Direct β-transitions from level a to level c in variant A, or from level b to level c in variant B, must be regarded as being of very low probability because the large spin difference between the initial and final states corresponds to an exceedingly low transition probability in the case of β-transitions.

Weizäcker[28] considered a further important question, namely, the production of isomeric nuclei. Let us illustrate Weizäcker's point of view with the example of rhodium. The ^{103}Rh (n,γ) reaction results in the formation of the compound nucleus ^{104}Rh* in a highly excited state (~7 MeV). This nucleus loses its excitation energy by successively emitting a series of γ-rays (dipole and quadrupole) and these γ-cascades proceed along different routes in different cases: in a certain fraction of ^{104}Rh* nuclei, the γ-cascade terminates in the ground state of ^{104}Rh and the "lower isomer" is produced; in all other ^{104}Rh* nuclei, the γ-cascade ends at a metastable level and the "upper isomer" ^{104m}Rh is produced.

1937–1941. The second part of a major review of what was known in nuclear physics was published in April 1937. The author of this second part, H. A. Bethe,[30] devoted a separate section to nuclear isomerism. He augmented Weizäcker's results by pointing out that the long lifetime of a metastable state did not necessarily signify that β-transition would proceed from this level: the excited state could be deexcited to the ground state of the same nucleus by the emission of a γ-ray (accompanied by internal conversion), i.e., by the transformation of the "upper isomer" into the "lower isomer" (now referred to as an isomeric transition). This can be illustrated by the scheme shown in Fig. 2.

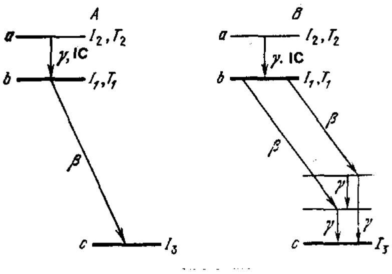

FIG. 2. Decay scheme for an isomeric nucleus (after Bethe). The notation is the same as in Fig. 1.

It is readily shown that, if a β-particle detector is used to record the β-ray intensity emitted by a given nuclide (N_β), and if $T_2 > T_1$, the $N_\beta(t)$ curve will consist of two components, corresponding to the two half-lives T_1 and T_2).11) Thus, the fact that two different β-transition half-lives have to be assigned to the same nuclide does not introduce further difficulty either in the case of parallel disintegration (Fig. 2). This was, in fact, stated (in passing) in Gamow's note.[11]

We note that there is a possible further, mixed, variant which was not noted by Bethe: one can have a branched disintegration from level a, so that a proportion of the nuclei belonging to the given nuclide decays via an isomeric transition and the remainder via a β-transition.

Since every nucleus has a set of excited states, Weizäcker's hypothesis clearly predicts that one can have instances of isomerism in which the metastable level belongs to a stable nucleus. The "lower isomer" is then a nonradioactive nucleus and the "upper isomer" emits γ-rays and conversion electrons of β-particles. This type of isomeric pair will have only one rather than two half-lives. This consequence of Weizäcker's hypothesis was pointed out independently by Bethe[30] and Pontecorvo.[31] Early experimental data on such isomers can be found in the review by Grinberg.[32]

We must now return to a further examination of the isomerism of bromine. Two papers were published in 1937, either of which would have been sufficient for a reliable assignment of the mass number to the isomeric nucleus of radiobromine. Bothe and Gentner undertook an investigation of photonuclear reactions using very hard γ-rays (~17 MeV) emitted when lithium is bombarded with protons.[33] They showed that the (γ,n) reaction occurred in all cases. Three radioactive bromine isotopes with half-lives of 5 ± 0.5 min, 16 ± 0.9 min, and 4.5 ± 0.1 h were produced when bromine was exposed to these γ-rays. Comparison of these results with previous data on the (n,γ) reaction showed incontrovertibly that the half-lives of 18 min and 4.5 h were those of ^{80}Br, the half-life of 5 min was that of ^{78}Br, and the half-life of 36 h was due to ^{82}Br.

Somewhat later, Snell[34] published a paper entirely devoted to the problem of the isomerism of bromine. He also used the method of "crossed nuclear reactions" but in a different version. He bombarded bromine and adjacent elements (As, Se, Kr, Rb) with deuterons and α-particles, accelerated in a cyclotron, and neutrons obtained from the (d + Be) reaction. He showed quite unambiguously that the activities with periods of 18.5 ± 0.5 min and 4.54 ± 0.10 h had to be ascribed to the isomeric nucleus ^{80}Br, whilst the activity with the half-life of 33.9 ± 0.3 h was that of ^{82}Br.

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11) The shape (and the end point) of the β-ray spectra is exactly the same for these two components since we are concerned with the same β-transition.

From the moment it was established that two half-lives had to be assigned to a particular radiobromine nucleus, namely, bromine-80, the Kurchatov hypothesis of nuclear isomerisom of bromine had to be regarded as confirmed experimentally.

The next step in the study of nuclear isomerism was, above all, to show experimentally that the general assumptions about the nature of this phenomenon, developed by Gamow and Weizäcker were, in fact, correct. The qeustions that had to be answered in the special case of bromine were at least three in number: (1) Which of the two varieties of bromine-80 was the excited isomer, (2) what was the excitation energy of the metastable state, and (3) was the decay scheme of these isomers of the series or the parallel type?

The first attempt to answer these questions was unsuccessful[34]: the author's conclusions were based on unconvincing arguments and turned out to be erroneous. The situation was completely clarified by the group working under the general direction of I. V. Kurchatov, whose "group leader" was L. I. Rusinov. In a carefully considered series of papers, they showed that the metastable state was ^{80}Br (4.5 h) and that this was the case of a series disintegration similar to that shown in Fig. 2 but somewhat more complicated than was implied by the theoretical descriptions of Weizäcker[28] and Bethe:[30] the metastable state was not the first but the second excited state of ^{80}Br with an excitation energy of about 86 keV. (Many examples of cascade isomeric transition were subsequently found.)

We shall now list the titles of these papers and comment upon them.

"The soft emission of bromine."[35] This paper was concerned with electrons of about 30 keV, detected among the radiations from ^{80}Br (4.5 h) and which, according to Kurchatov, had to be considered as conversion electrons.

"The emission of x rays by isomers of radioactive bromine."[36] Here, it was shown that radiobromine emitted x rays with a half-life of 4.5 h and that these were the characteristic x rays of bromine and not of krypton. This supported the hypothesis that the soft electrons from radiobromine were, in fact, conversion electrons, and indicated that this example of isomerism involved a series disintegration.

"Determination of the energy of conversion electrons from bromine isomers."[37] The energy was determined with a magnetic spectrometer, and the spectrum was found to contain *K* and *L* electrons. Their energies can be used to show that the corresponding transition energy was ~49 keV.

"The structure of the lower excited states of ^{80}Br."[38] Soft γ rays of 37 keV were found to be emitted with a half-life of 4.5 h, suggesting the presence of a two-stage transition from the metastable ^{80m}Br level to the ground state of this nucleus.

This detailed and systematic investigation of the isomerism of bromine was an important step forward, not only in solving particular problems, but also leading to a general understanding of the details of radioactive transformations of isomeric nuclei. The fact that all the results confirmed qualitatively the theoretical predictions with regard to the nature of nuclear isomerism was of major significance.

Since 1937, nuclear isomerism has been studied in many laboratories in different countries. As often happens in science generally, many of the ideas put forward for experiments of their interpretation evolved independently and almost simultaneously in different laboratories. For example, the conversion electrons emitted in the decay of metastable states were observed by the Kurchatov group in the case of bromine and by Pontecorvo in the case of rhodium.[39,40] Pontecorvo suggested[40] that the conversion origin of the emitted electrons could be demonstrated by looking for the characteristic x-ray spectrum. This was done independently by L. I. Rusinov and A. A. Yuzefovich[36] and by Abelson.[41] The determination of the spectrum of conversion electrons from bromine-80 was reported independently in two papers.[37,42] The fact that an isomeric transition (Fig. 2) was involved in the case of the bromine isomers was confirmed independently of the Leningrad group by the new and elegant method of "chemical separation of nuclear isomers."[43]

One of the main aims in the study of nuclear isomerism then became the experimental verification of the Weizäcker hypothesis concerning the reason for the metastability of the excited state. Direct demonstration of this involves measurement of the spins I_2 and I_1 of the metastable and ground state by one of the well-known methods (atomic beams, optical spectroscopy, and so on). Measurements of this kind rely on complex specialist equipment and the availability of a large amount of the radioactive material. Indirect methods were therefore used to determine ΔI.[44,45] They involved comparison between theoretical calculations and experimental data on quantities that were more or less rapidly varying functions of ΔI, namely, (1) the level lifetime, (2) the internal conversion coefficient α_c/α_L, $\alpha_{L_1}/\alpha_{L_2}$, and so on.

Data obtained by these methods confirmed the Weizäcker hypothesis. Direct measurements of the spins of the bromine isomers had to wait until 1959.[46] It was then shown that $I=5$ in the case of Br (4.5 h) and $I=1$ for Br (18 min). Figure 3 shows the level and transition scheme of ^{80}Br according to modern data.[47] Figure 4 shows currently available information[48] (in general outline) for ^{234}Pa, the first nuclear isomer to be discovered.

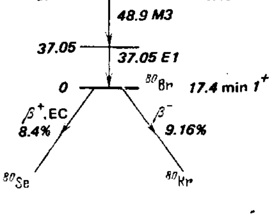

FIG. 3. Isomeric transition in ^{80}Br (EC = electron capture).

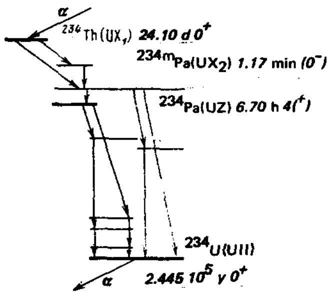

FIG. 4. Simplified level and transition scheme for ^{234}Th → → ^{234}Pa → ^{234}U. The β-ray spectrum of ^{234}Th consists of four components. The main component (72.5%) corresponds to the transition to ^{234m}Pa. The β-ray spectrum of ^{234}Pa consists of 20 (22?) components. 98.6% of all the β-transitions correspond to the transition to the ground state of ^{234}U. The excitation energy of ^{234m}Pa is about 74 keV.

Research performed at many laboratories showed quite early that the phenomenon of nuclear isomerism was not at all rare. Already by the end of 1940, there were 33 known cases of nuclear isomerism.[44] The continuous improvement in the experimental techniques of nuclear physics eventually resulted in the discovery and investigation of short-lived isomers. For example, many isomers were found in the millisecond range. It is now practically impossible to count the total number of known isomers without specifying some arbitrarily chosen boundary. The point is that we have reached a paradoxical situation: although the phrase, "metastable state" has now been used for many decades, there is as yet no agreement as to which levels are, in fact, metastable, i.e., what is the lower limit for the lifetime of this type of level. Lifetimes in the microsecond and nanosecond ranges have been measured for many nuclear states, for example, for the first excited state of ^{65}Zn (1.65 nsec; $E2$ transition, $\Delta E = 54$ keV). This level can hardly be regarded as isomeric.

The first few years of research into nuclear isomerism already yielded very extensive experimental data on the properties of nuclear states and the nature of transitions between them. These data were widely used to develop and verify various models in the theory of the nucleus. The above experimental methods for determining the type and order of the multipolarity of γ ray emission were developed as a result of studies of nuclear isomerism, and are now classic. They belong to the arsenal of techniques widely used in modern nuclear spectroscopy.

Later, still more extensive experimental data formed the basis for the discovery of many interesting regularities such as the existence of "islands of isomerism," which could be satisfactorily explained by the nuclear shell model, new types of isomerism such as "shape isomerism," "yrast traps," and so on. Research into nuclear isomerism is continuing.

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